



Substantial Mercury Releases and Local Deposition from Permafrost Peatland Wildfires in Southwestern Alaska

Citation

Scott Zolkos et al., "Substantial Mercury Releases and Local Deposition from Permafrost Peatland Wildfires in Southwestern Alaska," *Environmental Science & Technology* 58, no. 46 (2024): 20654–64, <https://doi.org/10.1021/acs.est.4c08765>.

Published version

<https://doi.org/10.1021/acs.est.4c08765>

Link

<https://dash.harvard.edu/handle/1/42730767>

Terms of use

This article was downloaded from Harvard University's DASH repository, and is made available under the terms and conditions applicable to Open Access Policy Articles (OAP), as set forth at

<https://harvardwiki.atlassian.net/wiki/external/NGY5NDE4ZjgzNTc5NDQzMGIzZWZhMGFIOWI2M2EwYTg>

Accessibility

<https://accessibility.huit.harvard.edu/digital-accessibility-policy>

Share Your Story

The Harvard community has made this article openly available.
Please share how this access benefits you. [Submit a story](#)

Substantial mercury releases and local deposition from permafrost peatland wildfires in southwestern Alaska

Scott Zolkos^{1,2*}, Benjamin M. Geyman¹, Stefano Potter², Michael Moubarak³, Brendan M. Rogers², Natalie Baillargeon², Sharmila Dey¹, Sarah M. Ludwig⁴, Sierra Melton⁵, Edauri Navarro-Pérez⁶, Ann McElvein^{7,8}, Prentiss H. Balcom¹, Susan M. Natali², Seeta Sistla⁹, Elsie M. Sunderland¹

Affiliations

¹Harvard John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge Massachusetts 02138, United States

²Woodwell Climate Research Center, Falmouth, Massachusetts 02540, United States

³Department of Biology, Hamilton College, Clinton, New York 13323, United States

⁴Department of Earth and Environmental Science, Columbia University, New York, New York 10027, United States

⁵Department of Geosciences and Earth and Environmental Systems Institute, Pennsylvania State University, University Park, Pennsylvania 16802, United States

⁶Program of Environmental Life Sciences, School of Life Sciences, Arizona State University, Tempe, Arizona 85281, United States

⁷University of California Berkeley, Berkeley California 94720, United States

⁸ICF International, Reston Virginia 20190, United States

⁹Department of Natural Resources Management and Environmental Sciences, California Polytechnic State University, San Luis Obispo, California 93407, United States

*e-mail: sgzolkos@gmail.com

Abstract. Increasing wildfire activity at northern high latitudes has the potential to mobilize large amounts of terrestrial mercury (Hg). However, understanding implications for Hg cycling and ecosystems is hindered by sparse research on peatland wildfire Hg emissions. In this study, we used measurements of soil organic carbon (SOC) and Hg, burn depth, and environmental indices derived from satellite remote sensing to develop machine learning models for predicting Hg emissions from major wildfires in permafrost peatland of the Yukon-Kuskokwim Delta (YKD) in southwestern Alaska. Wildfire Hg emissions during summer 2015 – estimated as the product of Hg:SOC (0.38 ± 0.17 ng Hg g C⁻¹), predicted SOC stores (mean [5th–95th] = 9.1 [5.3–11.2] kg C m⁻²), and burn depth (11.3 [8.2–13.9] cm) – were 556 [164–1138] kg Hg, or approximately 6% of Hg emissions from wildfire activity >60°N. Modeling estimates suggest that wildfire nearly doubled summertime Hg deposition within 10 kilometers, despite advection of more than 75% of total emissions beyond Alaska. YKD areal emissions combined with remote sensing estimates of burned area suggest that wildfire Hg emissions from northern peatlands (25.4 [14.9–33.6] Mg y⁻¹) are an important component of the northern Hg budget. Additional research is needed to refine these estimates and understand implications for Arctic and global Hg cycling.

Keywords: Hg, peat, emissions, transport, deposition, machine learning, uncertainty

Synopsis. Large stores of terrestrial mercury are vulnerable to increasing wildfire activity at northern high latitudes, yet few studies exist on northern peatland wildfire mercury release. This study estimates large releases and local deposition of mercury from peatland wildfires, with implications for the Arctic mercury budget.

1. INTRODUCTION

Mercury (Hg) is a naturally occurring toxic element that accumulates in food webs, posing a health risk to exposed humans.¹ Uptake and oxidation of gaseous elemental Hg (Hg^0) by vegetation and direct Hg^{II} deposition are thought to be the main inputs to terrestrial ecosystems ($\sim 2,300 \text{ Mg y}^{-1}$).^{2,3} Complexation of Hg^{II} by organic matter stabilizes it in soils and represents a main global sink of Hg.^{4,5} For millennia, a cold climate at northern high latitudes slowed the microbial decomposition of organic matter, promoting the accumulation of large amounts of Hg in upper soil layers ($26,000\text{--}72,000 \text{ Mg}$).^{6,7} Climate-driven warming and intensification of northern wildfire activity^{8,9} threatens this reservoir of terrestrial Hg.^{10,11} Early estimates suggested that annual Hg emissions from boreal wildfire ($23\text{--}53 \text{ Mg y}^{-1}$) were a major component of the northern ($>60^\circ\text{N}$) Hg budget.^{11,12} More recent budgets have suggested that Hg emissions from northern wildfire (8.8 Mg y^{-1})¹³ are minor relative to volatilization from terrestrial ecosystems (24 Mg y^{-1}), coastal erosion (39 Mg y^{-1})^{14,15}, riverine export (41 Mg y^{-1})^{16,17}, and annual dry and wet deposition to land (118 Mg y^{-1}).¹³ However, wildfire Hg emissions remain among the most uncertain flux terms in northern Hg budgets (e.g., refs 18–20), in part because of limited observational data from northern peatlands, where wildfires are thought to release up to several times more Hg per unit area compared to other ecosystem types.¹¹

Assessing peatland wildfire Hg emissions, transport, and deposition is essential for improving understanding of ecosystem Hg uptake. Hg emissions from a wildfire event are typically estimated by measuring atmospheric composition,^{12,18,20,21} or by assessing changes in soil Hg stores before and after fire.¹¹ Laboratory and field experiments can also help to constrain Hg emissions and speciation.^{19,22,23} Assessing wildfire Hg emissions at the regional or continental scale requires information on burned area and potential Hg release per unit area. These can be derived from observations or information on fuel load, fractional Hg loss (i.e., the proportion of fuel that experiences Hg release), and biome-specific emissions factors (the unit mass of Hg released per unit mass of fuel).^{13,24} Studies on peatland wildfire Hg emissions often assume complete release of Hg.^{19,20} Such an assumption is consistent with Hg release from soils within the temperature range of oxidation reactions observed in smoldering combustion that typify peat fire ($300\text{--}600^\circ\text{C}$).²⁵ Thus, uncertainties associated with Hg emissions at larger scales are primarily from burned area estimates,^{26–29} fuel load (i.e., soil Hg stores), and emissions factors.

Smoke plume height and Hg speciation influence the transport and deposition of wildfire Hg emissions.^{12,30} Particles and chemical constituents within the planetary boundary layer (PBL) are more likely to deposit at local to regional scales (10s to 100s km), whereas mixing into the free troposphere (FT) promotes hemispheric to global transport.^{31,32} Injection of plumes above the PBL requires convective energy, which depends on fire intensity and environmental conditions that influence fire behavior (e.g., moisture content, topography).^{31,33,34} The long residence time of Hg^0 in the atmosphere ($\sim 6\text{--}12$ months) facilitates its long-distance transport, whereas divalent inorganic Hg (Hg^{II}) species (including Hg^{II} sorbed to particles) deposit within days to weeks.³⁵ In peatlands, relatively high soil moisture reduces fuel availability and fire thermal energy, resulting in primarily smoldering combustion.³⁶ Consequently, peatland wildfires often have a relatively high particulate fraction, low convective energy, and smoke plumes that mostly remain within the PBL (e.g., $>80\%$ in boreal regions),^{31,37} with important implications for deposition to local and regional ecosystems. However, limited research on the release and fate of Hg from northern peatland wildfires^{11,23} hinders a more complete understanding of Hg cycling at northern high latitudes and ecosystem implications.

The main objective of this study was to empirically estimate Hg emissions from wildfires during summer 2015 in permafrost-affected tundra peatland in the Yukon-Kuskokwim Delta (YKD), Alaska, using measured Hg, SOC, and soil organic layer burn depth and environmental indices derived from satellite remote sensing data. We simulated Hg transport and deposition to local (YKD) and regional (Alaska) ecosystems using a global atmospheric chemical transport model (GEOS-Chem) forced by the range of wildfire Hg emissions estimated in our work and varying assumptions about smoke plume injection altitudes.^{31,37,38} We use estimates of IKU areal Hg emissions and burned area from a remote sensing product specific for northern high latitudes to scale wildfire emissions estimates to all northern peatlands, and consider implications for northern ecosystems.

2. METHODS

2.1. Study Area

The Izaviknek and Kingaglia Uplands (IKU) (61.3°N, 162.9°W) span 3900 km² within the YKD ecoregion. This rolling tundra landscape lies 10–65 m above sea level, and is characterized by peatland plateau-fen complexes and interconnected surface waters underlain by discontinuous permafrost³⁹ (Figure 1). Vegetation in unburned areas primarily consists of lichen, low shrubs, mosses, and sedges. Approximately 20% of the IKU (804 km²) has experienced wildfire since 1953, most of which occurred during three concurrent wildfires from late June to July, 2015 (443 km²)⁴⁰ (Table S1). Thus, the IKU offers an ideal environment to investigate peatland Hg release from wildfire. Wildfires in the IKU have relatively low severity due to moderate to high soil moisture. They typically combust surficial vegetation and organic matter in the top 15 cm of soil.^{39,41}

2.2. Burn Depth Measurements

During summer 2019, burn depth ($n = 832$) was measured at 107 transects between rafts of living moss that survived fire (Figure S1).⁴¹ Along each transect, burn depth was measured every 25 cm as the height between the burned soil surface and the pre-burn surface, which was reconstructed using a taught line between moss raft tops. While post-fire thaw-driven terrain subsidence (i.e., thermokarst) was not evident during fieldwork, measurements were not available to assess its potential effect on burn depth estimates.

2.3. Unburned Soil and Vegetation Collection and Processing

Soils from unburned plots were collected, processed, and analyzed for organic carbon (SOC) as detailed in ref 41. Soil cores 6 cm in diameter and ~30 cm long ($n = 126$) were collected in unburned areas from 2017 to 2019. Organics were separated into vegetation, fibric, and mesic layers following standard soil survey techniques.⁴² Because burn depth never exceeded the depth of the fibric layer,⁴¹ fibric material was homogenized, subsampled (~15 g), stored frozen, and used in analyses. Additional details are provided in the Supporting Information (Text S1).

Dominant plant functional types (PFT) were quantified every 1 m along thirteen 30 m transects at seven unburned sites using a point-intercept method.⁴³ At the end of each transect, aboveground biomass was harvested from plots either 0.09 m² (2018, $n = 12$) or 0.25 m² (2017, $n = 9$) and stored in separate paper bags for each PFT (lichen, evergreen shrub, deciduous shrub, moss, graminoid). Vegetation was later weighed and summed to determine plot-level aboveground biomass by PFT and in total.

2.4. Mercury and Carbon in Unburned Soils and Vegetation

Soils were dried (60°C, 48 h) and weighed for calculations of gravimetric moisture content (%) and bulk density (g cm^{-3}) at the Woodwell Climate Research Center. Oven-drying peat at 60°C may volatilize a small amount ($< 5\%$) of its Hg.⁴⁴ Soil organic matter was measured gravimetrically on combusted samples (450°C, 5 h), and total carbon (%) was analyzed using an elemental analyzer (Vario Max CN, Elementar). Vegetation was air dried, weighed for estimates of aboveground biomass, and ground for analysis of total carbon (LECO) and Hg.

Hg content in soil and vegetation was measured using a direct Hg analyzer (Nippon MA-3000) at Harvard University following published methods.⁴⁵ All sample boats were cleaned and blanked before use, and a sample duplicate, blank, and reference material (MESS-4 [National Research Council Canada] or NIST 1575a [U.S. Department of Commerce]) were analyzed every 10 samples. Blanks averaged $0.01 \text{ ng Hg g}^{-1}$ ($n = 43$). The mean coefficient of variation (CV) of duplicate Hg concentrations was 2.0% ($n = 40$), and concentrations of MESS-4 (mean $\pm$ SD = $79.5 \pm 1.3 \text{ ng g}^{-1}$, $n = 31$) and NIST 1575a (mean $\pm$ SD = $37.4 \pm 0.7 \text{ ng g}^{-1}$, $n = 16$) were within the reference value range.

Stores of soil Hg (mg m^{-2}) and SOC (kg m^{-2}) were calculated as the product of bulk density, fibric layer depth, and the content of Hg (ng g^{-1}) or organics (carbon or organic matter %)^{46,47} and scaled to a common depth of 30 cm assuming the fibric layer in shorter cores reached 30 cm. Measurements of soil Hg were averaged for samples collected along the same transects in unburned areas prior to modeling wildfire Hg emissions. Vegetation Hg stores ($\mu\text{g m}^{-2}$) were calculated as the summed products of plot-level PFT biomass (g m^{-2}) and average PFT Hg content (ng Hg g^{-1}), and averaged for transects at the same site. Samples collected from unburned areas in the IKU during 2017–2019 are taken to be representative of soils and vegetation pre-2015 wildfire for our Hg emissions model.

2.5. Wildfire Hg Emissions Modeling

2.5.1. Wildfire Hg Emissions and Uncertainty Analysis

Plot-level wildfire Hg emissions were estimated as the sum of soil Hg loss (pre-fire soil Hg stores $\times$ burn depth) and vegetation Hg loss (pre-fire vegetation Hg stores $\times$ fraction PFT combusted), assuming complete release of Hg.^{19,20} The fraction PFT combusted was derived from a study on post-fire vegetation change in the IKU³⁹ as the ratio of the coverage of each PFT ($f_{\text{PFT-}i}$) two years post-fire (C_b) to its coverage in unburned terrain (C_u): $f_{\text{PFT-}i} = 1 - (C_b / C_u)$. Pre-fire soil Hg stores (mg m^{-2}) in the top 30 cm of soil were calculated as the product of pre-fire SOC and the ratio of Hg:C (RHgC, $\mu\text{g Hg g C}^{-1}$) using our paired measurements of Hg and SOC ($n = 50$). Soil Hg stores are commonly estimated by scaling RHgC by an estimate of SOC stores.^{6,7,48}

To predict burn depth and pre-fire SOC stores for estimating soil Hg loss within the 2015 wildfire perimeters, we developed Random Forest regression models (detailed in next paragraph) from gridded geospatial data on environmental indices known to correlate with SOC and burn severity at northern high latitudes including vegetation greenness and health, water content, topography, and burn severity^{49,50} (Table S2). Indices were derived from cloud-free composites of Landsat 8 Collection 2 Tier 1 30 m-resolution global surface reflectance imagery⁵¹ collected from June 1–21, 2015 (pre-fire) or August 1–31, 2015 (post-fire). Indices on terrain morphology (elevation, slope, rugosity) were derived from a pre-fire 2 m-resolution digital surface model (ArcticDEM)⁵² for $>60^\circ\text{N}$ that we resampled to 30 m using bilinear interpolation. Perimeters of the 2015 wildfires were obtained from the Monitoring Trends in Burn Severity database for the United States.⁴⁰

We developed Random Forest models to predict burn depth and SOC stores using the *ranger* function in the R package *caret*^{53,54} following published methods.²⁸ We first excluded strongly correlated predictors ($-0.7 > r > 0.7$) using a Pearson correlogram. We then determined the optimal subset of remaining predictors using recursive feature elimination (RFE), which iteratively culls predictors by fitting a secondary Random Forest model that discards the least important features at each iteration.⁵⁵ We repeated the RFE 100 times across a leave-one-out cross validation (LOOCV) to determine the optimal subset of covariates. Next, we searched for the optimal Random Forest model parameters using the *train* function with a search grid of length 10, which tests every parameter combination and selects 10 random numbers per parameter for each test. Lastly, we evaluated the final fit of the Random Forest model using LOOCV repeated 100 times on the model with the optimal subset of predictors and model parameters. For both RFE and the full Random Forest model, LOOCV was used to maximize the proportion of our data on burn depth ($n = 106$) and SOC stores ($n = 51$) used for model calibration and withhold ample data for validation. Additional details on development of the Random Forest models are provided in the Supporting Information (Text S2).

We estimated wildfire Hg emissions and uncertainty for every 30 m grid cell within the 2015 wildfire perimeters ($n = 455,316$) using Monte Carlo simulations and resampling techniques for the five components of our emissions estimates: pre-fire soil and vegetation Hg stores, burn depth, and the fractions of soil and vegetation combusted. Uncertainties in each parameter were assumed to be random across grid cells and all simulations and resampling procedures were repeated 100 times each for every 30 m grid cell. For each simulation, inputs were predicted values (burn depth, SOC) with uncertainty from Random Forest LOOCV, or from the mean of observations (RHgC, pre-fire vegetation stores, fraction vegetation combusted) (Table S3). Our measurements on PFT Hg were used to estimate pre-fire vegetation Hg stores within the uncertainty analysis. The fraction of vegetation combusted for dominant PFTs was taken from observations on changes in PFT abundance two years after the 2015 IKU wildfires (see above).³⁹ Surface water was masked from outputs of SOC stores, burn depth, and Hg release using high-resolution waterbody polygons derived for the study area.⁵⁶ Hg release and uncertainty for each grid cell were calculated as areal release (m^{-2}) upscaled to the grid cell area (900 m^2), and then summed across all 455,316 cells to estimate total Hg emissions.

2.5.2. Atmospheric Transport and Deposition of Wildfire Hg Emissions

To assess the magnitude and distribution of Hg deposition from the IKU wildfires, we simulated atmospheric Hg deposition using a global 3-D atmospheric chemical transport model for Hg (GEOS-Chem, v.14.3).^{2,38} GEOS-Chem simulates the chemistry, transport, and deposition of atmospheric Hg species (Hg^0 , Hg^{II}).⁵⁷ The model has been extensively evaluated against available atmospheric observations.³⁸ Atmospheric transport is driven by assimilated meteorological data from the GEOS-FP product from the NASA Global Modeling and Assimilation Office.⁵⁸ The model was forced with 2015 global anthropogenic emissions of 2390 Mg y^{-1} , which included 69% released as Hg^0 .⁵⁹ Offline emissions from terrestrial ecosystems, ocean evasion, and geogenic emissions were held constant following values used in the most recent GEOS-Chem benchmark budget.³⁸

We first conducted global simulations at $2^\circ \times 2.5^\circ$ horizontal resolution with 47 vertical levels in order to establish boundary conditions for subsequent nested regional simulations at $0.25^\circ \times 0.3125^\circ$ in our modeling domain in southwest Alaska, which included portions of the watersheds of the Yukon and Kuskokwim Rivers. Total deposition refers to wet and dry deposition. Hg

transport and deposition were estimated using data on the fractions of total gaseous Hg (TGM) and total particulate Hg (TPM) in peat fire smoke (93.5–99.2% TGM)¹⁹ (Text S3, Table S4). We assumed that emitted TGM was entirely elemental Hg (Hg⁰) with negligible Hg^{II},²¹ and that TPM was entirely Hg^{II}. We assumed that 11% of emissions reach the FT based on results for boreal biomes in June from a 5-year survey of wildfire plume heights in North America.⁶⁰ Results are presented for three domains (Figure 1) representing the entire nested model grid centered on Alaska (51 – 73 °N, 138.5 – 170 °W; 3.7×10⁶ km²), the YKD (58 – 64 °N, 160 – 168 °W; 3.0×10⁵ km²), and the IKU fire region (61.25 – 61.5 °N, 162.5 – 163.125 °W; 2.8×10³ km²).

2.5.3. Estimating Northern Peatland Contributions to Wildfire Hg Emissions

To estimate peatland contributions to northern wildfire Hg emissions, we multiplied estimated areal peatland emissions and uncertainty (mg m⁻², Sec. 3.3) and boreal forest emissions (0.15 mg m⁻²)¹² by annual burned area (2002–2022) from a model specific to the tundra-boreal region (>50°N).²⁷ We separately calculated peatland and non-peatland burned area using the fractional distribution of peatlands⁶¹ associated with each burned pixel. While this MODIS-based product may underestimate burned area in northern peatlands,²⁶ our approach assumes that fractional peatland coverage in the larger (10 km resolution) pixels is the same at the scale of the burned area pixels (463 m), which may result in overestimation of peatland burned area, especially in pixels where peatlands are sparser.

2.6. Statistical Analyses

Geospatial data were processed using Google Earth Engine.⁶² ArcGIS Pro v.2.5. R software v.4.2.2 was used for all statistical analyses.⁶³ Random Forest models were developed using the *ranger* function in the R package *caret*.^{53,54} Ranges of estimated burn depth, SOC and Hg stores, and Hg emissions within the 5th and 95th percentiles of modeled estimates are presented as mean [5th–95th]. For Hg stores and emissions, the range is taken from the mean values for each pixel-wise Monte Carlo simulation across all pixels. Other values are reported as mean ± SD, unless noted.

3. RESULTS AND DISCUSSION

3.1. Terrestrial Hg in IKU Tundra Peatlands: Elevated Concentration, Intermediate Stores

Hg concentrations in unburned IKU soils (143 ± 62 ng g⁻¹) were elevated relative to organic soils elsewhere in the north (e.g., ca. 90–150 ng g⁻¹ in interior Alaska,⁷ 81 ± 57 ng g⁻¹ in Siberia⁶). The low bulk density of IKU soils (0.08 ± 0.05 g cm⁻³) was typical of less-humified, fibric upper layers in peat.^{6,7,64} These high Hg concentrations and low bulk densities account for moderate Hg stores of 3.3 ± 1.8 mg m⁻² in the upper 30 cm of IKU soils, which is similar to upper 30–40 cm peatland Hg stores in Siberia (0.3–6.0 mg m⁻²)⁶ and Sweden (0.9–4.7 mg m⁻²).^{64,65} Hg concentrations in the six most common PFTs in the IKU³⁹ were similar to those in other northern locations (Table S5). Lichen had intermediate Hg concentrations (47 ± 20 ng g⁻¹), and accounted for the most biomass (71%) and the largest aboveground Hg stores (65 ± 35 µg m⁻²). Hg stores in all vegetation PFTs (65 ± 25 µg m⁻², mean ± SD by site) were significantly lower than in soils (Welch's t-test with unequal variances: *p* < 0.001).

3.2. Burn Depth and Stores of SOC and Hg: Model Performance, Predictors, and Estimates

Random Forest models predicting burn depth and pre-fire SOC stores each had an *R*² of 0.25 across 100 iterations of LOOCV (Figure S2), demonstrating performance comparable to other burn

depth models (e.g., ref 28). LOOCV slopes < 1 indicated some underestimation, despite model optimization using recursive feature elimination and semi-automated model tuning procedures.^{28,55,66} This underestimation of burn depth and SOC stores, and thus perhaps Hg emissions, may be attributed to the size of the datasets and/or number of predictors.⁶⁷

Field measurements confirmed that burn depth (10.2 ± 3.5 cm) never exceeded the fibric layer depth (25.6 ± 6.6 cm).⁴¹ Predicted burn depth (11.3 [8.9–13.6] cm) aligned with estimates of burn depth for other northern high latitude peatlands (~ 5 –20 cm).^{68–74} Predicted burn depth was generally lower near lakes and ponds, and higher in terrains with less standing water (Figure S3). The burn depth model was most influenced by terrain surface variation (rugosity) and burn severity (dNBR) (Figure S4). A remote sensing index for terrestrial moisture content (TCW) had moderate influence, while slope had minimal influence yet was retained as a predictor (Table S6). dNBR and soil moisture are known to directly correlate with burn severity and combustion in northern ecosystems,^{50,75} while rugosity in the IKU generally varies between land cover classes that are associated with surface water (e.g., fens) and drier terrains (e.g., upland plateaus),⁷⁶ where burn depths were greater.

Predicted pre-fire carbon stores in the top 30 cm of organic soil (9.1 [5.9–11.0] kg m⁻²) within the 2015 wildfire perimeters were typically lower in proximity to water channels (Figure S5). The SOC stores model was most influenced by metrics which captured spectral information on vegetation greenness and health (TCG, NDVI), moderately influenced by elevation, and less influenced by terrestrial moisture content (Table S6). While direct correlations between SOC and remote sensing indices for vegetation productivity and terrestrial wetness commonly reflect photosynthesis and soil moisture as drivers of soil carbon accumulation,^{49,77,78} general anticorrelations between our measurements of SOC stores and these indices (Figure S6) reflect lower soil moisture and NDVI in lichen-dominated peat plateau uplands, where SOC stores can be high relative to riparian environments. Predicted pre-fire Hg stores (3.5 [2.2–4.3] mg m⁻²), derived by applying measurements of RHgC (0.38 ± 0.17 ng g⁻¹) to predictions of SOC, followed the spatial distribution of SOC stores.

3.3. Large Pulse of Hg Emissions from IKU Peatland Wildfire

Wildfire Hg emissions were 1.36 [0.8–1.8] mg m⁻² and were generally higher in uplands than in lower-elevation wetland areas (Figure 1). These emissions are comparable to sparse other estimates of northern wildfire Hg emissions in North American peatlands (1.6 – 7.0 mg m⁻²) and boreal forest (0.2 – 0.9 mg m⁻²) that are also based on post-fire soil Hg loss.^{11,23} Estimates for boreal regions based on emissions factors from top-down approaches (e.g., airborne smoke plume measurements, inversion models) are similar in magnitude (up to ~ 0.5 mg m⁻²), but mostly exclude peatlands.^{20,79} Summing the total emissions for each grid cell in our study area, we estimate that the IKU wildfires in 2015 released 556 [164–1138] kg Hg. These emissions were approximately $\sim 6\%$ of annual fire Hg emissions $>60^\circ\text{N}$.¹³ We estimate release of 130 $\mu\text{g Hg kg}^{-1}$ fuel from the IKU fires, calculated as mean Hg release (1.36 mg m⁻²) divided by fuel load (10.5 kg m⁻²). We estimated the latter as mean aboveground biomass (2.21 kg m⁻²) plus the product of mean bulk density (0.08 g cm⁻³) and burn depth (10.2 cm). This value is 0.9 – $1.3\times$ that of the values for emissions factors for boreal forest wildfire (103 to >140 $\mu\text{g kg}^{-1}$, depending on TPM fraction),^{18,20,79} and 0.2 – $0.7\times$ lower than emissions factors calculated for boreal peatland wildfire,¹¹ for which estimated Hg stores in the upper ~ 30 cm of soil were approximately five times higher than in this study. Refining the sparse and varied estimates of northern wildfire Hg emissions¹³

will benefit from efforts to constrain emissions factors and northern soil Hg stores, especially in peatlands and in more fire-vulnerable shallow soil layers and areas of drying.⁸⁰

3.4. Rapid Increase in Hg Deposition to Local IKU Ecosystems Attributed to Wildfire

Baseline total deposition from May 2015 to May 2016 was 49 kg over the IKU, 3.5 Mg over the YKD, and 42.7 Mg over Alaska (Table 1). The IKU wildfires contributed 30% [8–50%] of total Hg deposition to the IKU, 4% [1–9%] of deposition to the YKD, and ~1% of deposition to the Alaska domain during the active-fire period (June – July 2015), when the majority of deposition occurred (Figure 2, Figure 3). Of the 556 [164–1138] kg Hg released by the fires, 75 [16–160] kg was redeposited within the Alaskan domain. Per unit of emissions, 10% [10–11%] of TGM and 61% [58–65%] of TPM redeposited within the domain; the remainder was advected beyond the grid boundary. These patterns (i) emphasize pronounced local deposition of IKU peatland wildfire Hg, (ii) show relatively greater importance of TPM to the deposition budget nearby the wildfires, and (iii) indicate that most of the Hg released from the IKU wildfires was redistributed beyond Alaska.

Table 1 summarizes wildfire Hg emissions and deposition magnitudes across a range of scenarios which capture uncertainty in Hg emissions magnitude, TGM fraction, and the fraction of emissions lofted to the FT. Uncertainty in the magnitude of wildfire Hg emissions (Table S7) was the largest source of deposition uncertainty, with the lower-bound emission estimate producing a -71% change in deposition to all domains and the upper-bound estimate producing +105% change relative to the central emission estimate. The second largest source of uncertainty was the particle-associated fraction of Hg emissions (Figure S7). Under the scenario in which 0.8% of wildfire Hg emissions are released as TPM (99.2% TGM), deposition is 22% lower over the Alaska domain and 29–30% lower over the IKU and YKD domains. Uncertainty in plume injection altitude was a relatively small source of uncertainty, with 5–7% lower deposition across all domains in the higher plume injection altitude case (18% to FT) and 4–6% higher deposition across all domains in the lower plume injection altitude case (5% to FT). Deposition is 9–11% higher under a scenario in which all fire-associated Hg emissions are released to the PBL, as compared with the central estimate (11% to FT).

Simulated deposition and concentrations compare favorably to available regional observations (Figure S8). Normalized mean bias (NMB) was 6% and root mean squared error (RMSE) was 0.14 ng m⁻³ ($n = 2$) for annual-average GEM concentration measurements from AMNet sites. NMB was 17% and RMSE was 1.8 µg m⁻² y⁻¹ ($n = 5$) for annual-average Hg wet deposition from sites in the Mercury Deposition Network (MDN) (Figure S8). Compared with the only annual dry deposition estimate available for the region, NMB was -18% and RMSE was 1.65 µg m⁻² y⁻¹. Simulated baseline rates of total deposition to land surfaces were higher over the IKU and YKD (17.8 µg m⁻² y⁻¹) than the Alaska domain average (13.9 µg m⁻² y⁻¹). Increasing atmospheric Hg monitoring across tundra and boreal North America, where there is currently a sparsity of active sites,⁸¹ will bolster future efforts to constrain Hg emissions and deposition as northern wildfire activity intensifies.

3.5. Implications for Hg Cycling at Northern High Latitudes

Our Hg emission estimates and simulated patterns of deposition show that northern peatland wildfire effects on Hg cycling and ecosystems are scale-dependent. Locally, soil Hg loss from the IKU wildfires was likely orders of magnitude greater than annual Hg deposition (e.g., ~4–10 µg m⁻² y⁻¹ in Alaska and Sweden).^{5,64} Hence, peatland wildfire, while brief, affects the annual and

perhaps decadal Hg mass budget, shifting burned terrain to a net Hg source through emissions alone. Wildfire-induced permafrost thaw may trigger indirect impacts on Hg cycling not captured by this study. For instance, active layer deepening in years to decades after fire³⁹ may increase the availability of permafrost Hg for hydrologic export to aquatic ecosystems. Over decades to centuries, post-fire trajectories of ecosystem recovery will shape the legacy of wildfire impacts on local Hg cycling. For example, vegetation community restructuring⁸² and uptake of Hg⁰ by successional vegetation,⁵ repletion of peatland Hg stores,⁸³ the frequency and severity of additional wildfire activity,⁸⁴ and wildfire- and thaw-induced changes to local ecohydrology (e.g., terrain drying or wetting) are all likely to influence post-fire Hg cycling.⁸⁵ Our finding that IKU peatland wildfire affects local Hg cycling further motivates the question arising from earlier research (e.g., refs 11,79): What is the potential magnitude of Hg emissions from all peatland wildfires across the north?

Multiplying our areal peatland wildfire Hg emissions and uncertainty by estimates of mean annual peatland burned area (2002-2022) from a tundra-boreal-specific fire model²⁷ and peatland distribution⁶¹ allows for a first estimate of soil and understory wildfire Hg emissions from northern peatlands (25.4 [14.9–33.6] Mg y⁻¹) and contribution to total northern wildfire Hg emissions (50 [29–66]%) (Figure 4). This estimate reflects variation in environmental conditions that influence peatland Hg stores and susceptibility to combustion across the IKU study region, but not necessarily in northern peatlands elsewhere.^{11,50} Nevertheless, we suggest that Hg emissions from northern peatland wildfire may be larger than estimated¹³ and thus a more important source of Arctic and global Hg pollution than is currently understood.⁸⁶ Improved understanding of the magnitude and fate of northern wildfire Hg release will require additional research to determine controls on the susceptibility of peatland Hg stores to combustion,¹⁹ measure variation of Hg speciation in smoke plumes from northern wildfires across diverse environmental conditions,⁸⁶ and reconcile differences in emissions factors estimated from ‘bottom-up’ field-based mass balance approaches (e.g., ref 11) versus ‘top-down’ aircraft measurements (e.g., ref 20) or inversion techniques (Sec. 3.3). To upscale estimates of wildfire Hg release, these complimentary approaches require robust estimates of burned area and fuel consumption that are typically derived from satellite remote sensing data. However, the relatively small area and low intensity of peat fires can challenge detection by satellite remote sensing platforms (e.g., MODIS) that have traditionally informed burned area products.^{87–89} Next-generation satellites that are designed to detect wildfire, such as WildFireSat (nominal deployment in 2029),⁹⁰ will be important tools for understanding northern fire regimes and Hg cycling.

In this study, we found that permafrost peatland wildfires in southwest Alaska released a large amount of terrestrial Hg and that rapid deposition contributed significantly to local Hg budgets, adding to scarce research on the magnitude and fate of northern wildfire Hg emissions.^{11,12,18,20,23,86} As climate warming increases tundra and boreal wildfire activity,^{8,9} we emphasize the need for new measurements of Hg release and improved wildfire monitoring capabilities, especially in peatlands, to refine understanding of wildfire effects on Hg cycling at northern high latitudes.¹⁰

ASSOCIATED CONTENT

Supporting Information

The supporting information is available free of charge at [URL TBD]: Field and laboratory methods (Text S1), Random Forest model development (Text S2), atmospheric transport and deposition of wildfire Hg emissions (Text S3), metadata on wildfires in the study area (Table S1), details on remotely sensed indices used in developing Random Forest models (Table S2),

constraints on inputs to wildfire Hg emissions estimates and uncertainty (Table S3), synthesis of published observations on Hg speciation in wildfire smoke plumes (Table S4), summary statistics on Hg and C content for soils and vegetation (Table S5), variables selected for Random Forest models of soil organic carbon and burn depth (Table S6), sensitivity of simulated deposition to uncertainty in emission characteristics (Table S7), map of field sites (Figure S1), performance metrics for Random Forest models (Figure S2), map of burn depth predicted by the Random Forest model (Figure S3), partial dependences of predictors in the Random Forest model of burn depth (Figure S4), map of soil organic carbon stores predicted by the Random Forest model (Figure S5), partial dependences of predictors in the Random Forest model of soil organic carbon (Figure S6), Hg deposition and uncertainties in the study domain (Figure S7), GEOS-Chem model evaluation (Figure S8).

Supporting code can be downloaded from <http://github.com/szolkos/ykd-wildfire-hg/>.
Supporting data can be downloaded from <https://doi.org/10.7910/DVN/ZA5JBZ>.⁹¹

AUTHOR INFORMATION

Corresponding Author

Scott Zolkos – *Harvard John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge Massachusetts 02138, United States & Woodwell Climate Research Center, Falmouth Massachusetts 02540, United States*

Authors

Benjamin M. Geyman – *Harvard John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge Massachusetts 02138, United States*

Stefano Potter – *Woodwell Climate Research Center, Falmouth Massachusetts 02540, United States*

Michael Moubarak – *Department of Biology, Hamilton College, Clinton New York 13323, United States*

Brendan M. Rogers – *Woodwell Climate Research Center, Falmouth Massachusetts 02540, United States*

Natalie Baillargeon – *Woodwell Climate Research Center, Falmouth Massachusetts 02540, United States*

Sharmila Dey – *Harvard John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge Massachusetts 02138, United States*

Sarah M. Ludwig – *Department of Earth and Environmental Science, Columbia University, New York, New York 10027, United States*

Sierra Melton – *Department of Geosciences and Earth and Environmental Systems Institute, Pennsylvania State University, University Park, Pennsylvania 16802, United States*

Edauri Navarro – *Program of Environmental Life Sciences, School of Life Sciences, Arizona State University, Tempe Arizona 85281, United States*

Ann McElvein – *University of California Berkeley, Berkeley California 94720, United States & ICF International, Reston Virginia 20190, United States*

Prentiss H. Balcom – *Harvard John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge Massachusetts 02138, United States*

Susan M. Natali – *Woodwell Climate Research Center, Falmouth Massachusetts 02540, United States*

Seeta Sistla – *Department of Natural Resources and Environmental Sciences, California Polytechnic State University, San Luis Obispo California 93407, United States*

Elsie M. Sunderland – *Harvard John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge Massachusetts 02138, United States*

Notes

The authors declare no competing financial interests.

ACKNOWLEDGEMENTS

This material is based upon work supported by the National Science Foundation under Award No. 1952715 and 2210173. Any opinions, findings, and conclusions or recommendations expressed in this material are those of the authors and do not necessarily reflect the views of the National Science Foundation.

REFERENCES

- (1) AMAP. *AMAP Assessment 2011: Mercury in the Arctic*; Arctic Monitoring and Assessment Programme (AMAP): Oslo, Norway, 2011; p 193.
- (2) Feinberg, A.; Dlamini, T.; Jiskra, M.; Shah, V.; Selin, N. E. Evaluating Atmospheric Mercury (Hg) Uptake by Vegetation in a Chemistry-Transport Model. *Environ. Sci. Process. Impacts* **2022**, 24 (9), 1303–1318. <https://doi.org/10.1039/D2EM00032F>.
- (3) Zhou, J.; Obrist, D.; Dastoor, A.; Jiskra, M.; Ryjkov, A. Vegetation Uptake of Mercury and Impacts on Global Cycling. *Nat. Rev. Earth Environ.* **2021**, 2, 269–284. <https://doi.org/10.1038/s43017-021-00146-y>.
- (4) Jiskra, M.; Wiederhold, J. G.; Skjellberg, U.; Kronberg, R.-M.; Kretzschmar, R. Source Tracing of Natural Organic Matter Bound Mercury in Boreal Forest Runoff with Mercury Stable Isotopes. *Environ. Sci. Process. Impacts* **2017**, 19 (10), 1235–1248. <https://doi.org/10.1039/C7EM00245A>.
- (5) Obrist, D.; Agnan, Y.; Jiskra, M.; Olson, C. L.; Colegrove, D. P.; Hueber, J.; Moore, C. W.; Sonke, J. E.; Helmig, D. Tundra Uptake of Atmospheric Elemental Mercury Drives Arctic Mercury Pollution. *Nature* **2017**, 547 (7662), 201–204. <https://doi.org/10.1038/nature22997>.

- (6) Lim, A. G.; Jiskra, M.; Sonke, J. E.; Loiko, S. V.; Kosykh, N.; Pokrovsky, O. S. A Revised Pan-Arctic Permafrost Soil Hg Pool Based on Western Siberian Peat Hg and Carbon Observations. *Biogeosciences* **2020**, *17* (12), 3083–3097. <https://doi.org/10.5194/bg-17-3083-2020>.
- (7) Olson, C.; Jiskra, M.; Biester, H.; Chow, J.; Obrist, D. Mercury in Active-Layer Tundra Soils of Alaska: Concentrations, Pools, Origins, and Spatial Distribution. *Glob. Biogeochem. Cycles* **2018**, *32* (7), 1058–1073. <https://doi.org/10.1029/2017GB005840>.
- (8) Descals, A.; Gaveau, D. L. A.; Verger, A.; Sheil, D.; Naito, D.; Peñuelas, J. Unprecedented Fire Activity above the Arctic Circle Linked to Rising Temperatures. *Science* **2022**, *378* (6619), 532–537. <https://doi.org/10.1126/science.abn9768>.
- (9) Hu, F. S.; Higuera, P. E.; Duffy, P.; Chipman, M. L.; Rocha, A. V.; Young, A. M.; Kelly, R.; Dietze, M. C. Arctic Tundra Fires: Natural Variability and Responses to Climate Change. *Front. Ecol. Environ.* **2015**, *13* (7), 369–377. <https://doi.org/10.1890/150063>.
- (10) McCarter, C. P. R.; Clay, G. D.; Wilkinson, S. L.; Page, S.; Shuttleworth, E. L.; Davidson, S. J.; Taufik, M.; Sigmund, G.; Waddington, J. M. Peat Fires and the Unknown Risk of Legacy Metal and Metalloid Pollution. *Environ. Res. Lett.* **2023**, *18* (7), 071003. <https://doi.org/10.1088/1748-9326/acddfc>.
- (11) Turetsky, M. R.; Harden, J. W.; Friedli, H. R.; Flannigan, M.; Payne, N.; Crock, J.; Radke, L. Wildfires Threaten Mercury Stocks in Northern Soils. *Geophys. Res. Lett.* **2006**, *33* (16), L16403. <https://doi.org/10.1029/2005GL025595>.
- (12) Sigler, J. M.; Lee, X.; Munger, W. Emission and Long-Range Transport of Gaseous Mercury from a Large-Scale Canadian Boreal Forest Fire. *Environ. Sci. Technol.* **2003**, *37* (19), 4343–4347. <https://doi.org/10.1021/es026401r>.
- (13) Dastoor, A.; Angot, H.; Bieser, J.; Christensen, J. H.; Douglas, T. A.; Heimbürger-Boavida, L.-E.; Jiskra, M.; Mason, R. P.; McLagan, D. S.; Obrist, D.; Outridge, P. M.; Petrova, M. V.; Ryjkov, A.; St. Pierre, K. A.; Schartup, A. T.; Soerensen, A. L.; Toyota, K.; Travnikov, O.; Wilson, S. J.; Zdanowicz, C. Arctic Mercury Cycling. *Nat. Rev. Earth Environ.* **2022**. <https://doi.org/10.1038/s43017-022-00269-w>.
- (14) Soerensen, A. L.; Jacob, D. J.; Schartup, A. T.; Fisher, J. A.; Lehnher, I.; St. Louis, V. L.; Heimbürger, L.-E.; Sonke, J. E.; Krabbenhoft, D. P.; Sunderland, E. M. A Mass Budget for Mercury and Methylmercury in the Arctic Ocean. *Glob. Biogeochem. Cycles* **2016**, *30* (4), 560–575. <https://doi.org/10.1002/2015GB005280>.
- (15) Fisher, J. A.; Jacob, D. J.; Soerensen, A. L.; Amos, H. M.; Steffen, A.; Sunderland, E. M. Riverine Source of Arctic Ocean Mercury Inferred from Atmospheric Observations. *Nat. Geosci.* **2012**, *5* (7), 499–504. <https://doi.org/10.1038/ngeo1478>.
- (16) Sonke, J. E.; Teisserenc, R.; Heimbürger-Boavida, L.-E.; Petrova, M. V.; Maruszczak, 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.* **2018**, *115* (50), E11586–E11594. <https://doi.org/10.1073/pnas.1811957115>.
- (17) 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* (7), 4140–4148. <https://doi.org/10.1021/acs.est.9b07145>.
- (18) Friedli, H. R.; Radke, L. F.; Lu, J. Y.; Banic, C. M.; Eisner, W. R.; MacPherson, J. I. Mercury Emissions from Burning of Biomass from Temperate North American Forests:

- Laboratory and Airborne Measurements. *Atmos. Environ.* **2003**, 37 (2), 253–267.
[https://doi.org/10.1016/S1352-2310\(02\)00819-1](https://doi.org/10.1016/S1352-2310(02)00819-1).
- (19) Kohlenberg, A. J.; Turetsky, M. R.; Thompson, D. K.; Branfireun, B. A.; Mitchell, C. P. J. Controls on Boreal Peat Combustion and Resulting Emissions of Carbon and Mercury. *Environ. Res. Lett.* **2018**, 13 (3), 035005. <https://doi.org/10.1088/1748-9326/aa9ea8>.
- (20) McLagan, D. S.; Stupple, G. W.; Darlington, A.; Hayden, K.; Steffen, A. Where There Is Smoke There Is Mercury: Assessing Boreal Forest Fire Mercury Emissions Using Aircraft and Highlighting Uncertainties Associated with Upscaling Emissions Estimates. *Atmospheric Chem. Phys.* **2021**, 21, 5635–5653. <https://doi.org/10.5194/acp-2020-1119>.
- (21) Wang, Y.; Huang, J.; Znananski, T. J.; Hopke, P. K.; Holsen, T. M. Impacts of the Canadian Forest Fires on Atmospheric Mercury and Carbonaceous Particles in Northern New York. *Environ. Sci. Technol.* **2010**, 44 (22), 8435–8440. <https://doi.org/10.1021/es1024806>.
- (22) Friedli, H. R.; Radke, L. F.; Lu, J. Y. Mercury in Smoke from Biomass Fires. *Geophys. Res. Lett.* **2001**, 28 (17), 3223–3226. <https://doi.org/10.1029/2000GL012704>.
- (23) Harden, J. W.; Neff, J. C.; Sandberg, D. V.; Turetsky, M. R.; Ottmar, R.; Gleixner, G.; Fries, T. L.; Manies, K. L. Chemistry of Burning the Forest Floor during the FROSTFIRE Experimental Burn, Interior Alaska, 1999. *Glob. Biogeochem. Cycles* **2004**, 18 (3), n/a-n/a. <https://doi.org/10.1029/2003GB002194>.
- (24) Kumar, A.; Wu, S.; Huang, Y.; Liao, H.; Kaplan, J. O. Mercury from Wildfires: Global Emission Inventories and Sensitivity to 2000–2050 Global Change. *Atmos. Environ.* **2018**, 173, 6–15. <https://doi.org/10.1016/j.atmosenv.2017.10.061>.
- (25) Rein, G.; Cleaver, N.; Ashton, C.; Pironi, P.; Torero, J. L. The Severity of Smouldering Peat Fires and Damage to the Forest Soil. *CATENA* **2008**, 74 (3), 304–309. <https://doi.org/10.1016/j.catena.2008.05.008>.
- (26) Blackford, K. R.; Kasoar, M.; Burton, C.; Burke, E.; Prentice, I. C.; Voulgarakis, A. INFERNO-Peat v1.0.0: A Representation of Northern High-Latitude Peat Fires in the JULES-INFERNO Global Fire Model. *Geosci. Model Dev.* **2024**, 17 (8), 3063–3079. <https://doi.org/10.5194/gmd-17-3063-2024>.
- (27) Loboda, T. V.; Hall, J. V.; Chen, D.; Hoffman-Hall, A.; Shevade, V. S.; Argueta, F.; Liang, X. Arctic Boreal Annual Burned Area, Circumpolar Boreal Forest and Tundra, V2, 2002–2022. **2024**. <https://doi.org/10.3334/ORNLDAAAC/2328>.
- (28) Potter, S.; Cooperdock, S.; Veraverbeke, S.; Walker, X.; Mack, M. C.; Goetz, S. J.; Baltzer, J.; Bourgeau-Chavez, L.; Burrell, A.; Dieleman, C.; French, N.; Hantson, S.; Hoy, E. E.; Jenkins, L.; Johnstone, J. F.; Kane, E. S.; Natali, S. M.; Randerson, J. T.; Turetsky, M. R.; Whitman, E.; Wiggins, E.; Rogers, B. M. Burned Area and Carbon Emissions across Northwestern Boreal North America from 2001–2019. *Biogeosciences* **2023**, 20 (13), 2785–2804. <https://doi.org/10.5194/bg-20-2785-2023>.
- (29) van Wees, D.; van der Werf, G. R.; Randerson, J. T.; Rogers, B. M.; Chen, Y.; Veraverbeke, S.; Giglio, L.; Morton, D. C. Global Biomass Burning Fuel Consumption and Emissions at 500 m Spatial Resolution Based on the Global Fire Emissions Database (GFED). *Geosci. Model Dev.* **2022**, 15 (22), 8411–8437. <https://doi.org/10.5194/gmd-15-8411-2022>.
- (30) Carpi, A. Mercury from Combustion Sources: A Review of the Chemical Species Emitted and Their Transport in the Atmosphere. *Water. Air. Soil Pollut.* **1997**, 98 (3–4), 241–254. <https://doi.org/10.1007/BF02047037>.

- (31) Val Martin, M.; Logan, J. A.; Kahn, R. A.; Leung, F.-Y.; Nelson, D. L.; Diner, D. J. Smoke Injection Heights from Fires in North America: Analysis of 5 Years of Satellite Observations. *Atmos Chem Phys* **2010**, *10* (4), 1491–1510. <https://doi.org/10.5194/acp-10-1491-2010>.
- (32) Geyman, B. M.; Thackray, C. P.; Jacob, D. J.; Sunderland, E. M. Impacts of Volcanic Emissions on the Global Biogeochemical Mercury Cycle: Insights From Satellite Observations and Chemical Transport Modeling. *Geophys. Res. Lett.* **2023**, *50* (21), e2023GL104667. <https://doi.org/10.1029/2023GL104667>.
- (33) Rogers, B. M.; Soja, A. J.; Goulden, M. L.; Randerson, J. T. Influence of Tree Species on Continental Differences in Boreal Fires and Climate Feedbacks. *Nat. Geosci.* **2015**, *8* (3), 228–234. <https://doi.org/10.1038/ngeo2352>.
- (34) Turquety, S.; Logan, J. A.; Jacob, D. J.; Hudman, R. C.; Leung, F. Y.; Heald, C. L.; Yantosca, R. M.; Wu, S.; Emmons, L. K.; Edwards, D. P.; Sachse, G. W. Inventory of Boreal Fire Emissions for North America in 2004: Importance of Peat Burning and Pyroconvective Injection. *J. Geophys. Res. Atmospheres* **2007**, *112* (D12), 2006JD007281. <https://doi.org/10.1029/2006JD007281>.
- (35) Horowitz, H. M.; Jacob, D. J.; Zhang, Y.; Dibble, T. S.; Slemr, F.; Amos, H. M.; Schmidt, J. A.; Corbitt, E. S.; Marais, E. A.; Sunderland, E. M. A New Mechanism for Atmospheric Mercury Redox Chemistry: Implications for the Global Mercury Budget. *Atmospheric Chem. Phys.* **2017**, *17* (10), 6353–6371. <https://doi.org/10.5194/acp-17-6353-2017>.
- (36) Rein, G.; Huang, X. Smouldering Wildfires in Peatlands, Forests and the Arctic: Challenges and Perspectives. *Curr. Opin. Environ. Sci. Health* **2021**, *24*, 100296. <https://doi.org/10.1016/j.coesh.2021.100296>.
- (37) Kahn, R. A.; Chen, Y.; Nelson, D. L.; Leung, F.; Li, Q.; Diner, D. J.; Logan, J. A. Wildfire Smoke Injection Heights: Two Perspectives from Space. *Geophys. Res. Lett.* **2008**, *35* (4), 2007GL032165. <https://doi.org/10.1029/2007GL032165>.
- (38) Shah, V.; Jacob, D. J.; Thackray, C. P.; Wang, X.; Sunderland, E. M.; Dibble, T. S.; Saiz-Lopez, A.; Černušák, I.; Kellö, V.; Castro, P. J.; Wu, R.; Wang, C. Improved Mechanistic Model of the Atmospheric Redox Chemistry of Mercury. *Environ. Sci. Technol.* **2021**, *55* (21), 14445–14456. <https://doi.org/10.1021/acs.est.1c03160>.
- (39) Frost, G. V.; Loehman, R. A.; Saperstein, L. B.; Macander, M. J.; Nelson, P. R.; Paradis, D. P.; Natali, S. M. Multi-Decadal Patterns of Vegetation Succession after Tundra Fire on the Yukon-Kuskokwim Delta, Alaska. *Environ. Res. Lett.* **2020**, *15* (2), 025003. <https://doi.org/10.1088/1748-9326/ab5f49>.
- (40) Eidenshink, J.; Schwind, B.; Brewer, K.; Zhu, Z.-L.; Quayle, B.; Howard, S. A Project for Monitoring Trends in Burn Severity. *Fire Ecol.* **2007**, *3* (1), 3–21. <https://doi.org/10.4996/fireecology.0301003>.
- (41) Moubarak, M.; Sistla, S.; Potter, S.; Natali, S. M.; Rogers, B. M. Carbon Emissions and Radiative Forcings from Tundra Wildfires in the Yukon-Kuskokwim River Delta, Alaska. *Biogeosciences* **2023**, *20*, 1537–1557. <https://doi.org/10.5194/bg-2022-144>.
- (42) Manies, K.; Waldrop, M.; Harden, J. Generalized Models to Estimate Carbon and Nitrogen Stocks of Organic Soil Horizons in Interior Alaska. *Earth Syst. Sci. Data* **2020**, *12* (3), 1745–1757. <https://doi.org/10.5194/essd-12-1745-2020>.
- (43) Karl, J. W.; McCord, S. E.; Hadley, B. C. A Comparison of Cover Calculation Techniques for Relating Point-Intercept Vegetation Sampling to Remote Sensing Imagery. *Ecol. Indic.* **2017**, *73*, 156–165. <https://doi.org/10.1016/j.ecolind.2016.09.034>.

- (44) Smeds, J.; Öquist, M.; Nilsson, M. B.; Bishop, K. A Simplified Drying Procedure for Analysing Hg Concentrations. *Water. Air. Soil Pollut.* **2022**, *233* (6), 216. <https://doi.org/10.1007/s11270-022-05678-7>.
- (45) Li, M.; Schartup, A. T.; Valberg, A. P.; Ewald, J. D.; Krabbenhoft, D. P.; Yin, R.; Balcom, P. H.; Sunderland, E. M. Environmental Origins of Methylmercury Accumulated in Subarctic Estuarine Fish Indicated by Mercury Stable Isotopes. *Environ. Sci. Technol.* **2016**, *50* (21), 11559–11568. <https://doi.org/10.1021/acs.est.6b03206>.
- (46) Ludwig, S.; Holmes, R. M.; Natali, S.; Schade, J.; Mann, P. Yukon-Kuskokwim Delta Fire: Soil and Permafrost Carbon, Nitrogen, Horizons, and Ice Structure, Alaska, 2016, 2018. <https://doi.org/10.18739/A2TQ5RD9K>.
- (47) Ludwig, S.; Holmes, R. M.; Natali, S.; Mann, P.; Schade, J.; Jardine, L.; Melton, S.; Navarro-Perez, E. Polaris Project 2017: Soil Fluxes, Carbon, and Nitrogen, Yukon-Kuskokwim Delta, Alaska, 2018. <https://doi.org/10.18739/A2Q23R08G>.
- (48) 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* (3), 1463–1471. <https://doi.org/10.1002/2017GL075571>.
- (49) Siewert, M. B. High-Resolution Digital Mapping of Soil Organic Carbon in Permafrost Terrain Using Machine Learning: A Case Study in a Sub-Arctic Peatland Environment. *Biogeosciences* **2018**, *15* (6), 1663–1682. <https://doi.org/10.5194/bg-15-1663-2018>.
- (50) Walker, X. J.; Rogers, B. M.; Veraverbeke, S.; Johnstone, J. F.; Baltzer, J. L.; Barrett, K.; Bourgeau-Chavez, L.; Day, N. J.; de Groot, W. J.; Dieleman, C. M.; Goetz, S.; Hoy, E.; Jenkins, L. K.; Kane, E. S.; Parisien, M.-A.; Potter, S.; Schuur, E. A. G.; Turetsky, M.; Whitman, E.; Mack, M. C. Fuel Availability Not Fire Weather Controls Boreal Wildfire Severity and Carbon Emissions. *Nat. Clim. Change* **2020**, *10*, 1130–1136. <https://doi.org/10.1038/s41558-020-00920-8>.
- (51) Crawford, C. J.; Roy, D. P.; Arab, S.; Barnes, C.; Vermote, E.; Hulley, G.; Gerace, A.; Choate, M.; Engebretson, C.; Micijevic, E.; Schmidt, G.; Anderson, C.; Anderson, M.; Bouchard, M.; Cook, B.; Dittmeier, R.; Howard, D.; Jenkerson, C.; Kim, M.; Kleyians, T.; Maersperger, T.; Mueller, C.; Neigh, C.; Owen, L.; Page, B.; Pahlevan, N.; Rengarajan, R.; Roger, J.-C.; Sayler, K.; Scaramuzza, P.; Skakun, S.; Yan, L.; Zhang, H. K.; Zhu, Z.; Zahn, S. The 50-Year Landsat Collection 2 Archive. *Sci. Remote Sens.* **2023**, *8*, 100103. <https://doi.org/10.1016/j.srs.2023.100103>.
- (52) Porter, C.; Morin, P.; Howat, I.; Noh, M.-J.; Bates, B.; Peterman, K.; Keesey, S.; Schlenk, M.; Gardiner, J.; Tomko, K.; Willis, M.; Kelleher, C.; Cloutier, M.; Husby, E.; Foga, S.; Nakamura, H.; Platson, M.; Wethington Jr., M.; Williamson, C.; Bauer, G.; Enos, J.; Arnold, G.; Kramer, W.; Becker, P.; Doshi, A.; D’Souza, C.; Cummins, P.; Laurier, F.; Bojesen, M. *ArcticDEM*. Harvard Dataverse, V1. <https://doi.org/10.7910/DVN/OHHUKH>.
- (53) Kuhn, M.; Wing, J.; Weston, S.; Williams, A.; Keefer, C.; Engelhardt, A.; Cooper, T.; Mayer, Z.; Kenkel, B.; R Core Team; Benesty, M.; Lescarbeau, R.; Ziem, A.; Scrucca, L.; Tang, Y.; Candan, C.; Hunt, T. R Package “Caret,” 2022. <https://cran.r-project.org/web/packages/caret/index.html>.
- (54) Wright, M. N.; Ziegler, A. **Ranger** : A Fast Implementation of Random Forests for High Dimensional Data in C++ and R. *J. Stat. Softw.* **2017**, *77* (1). <https://doi.org/10.18637/jss.v077.i01>.

- (55) Darst, B. F.; Malecki, K. C.; Engelman, C. D. Using Recursive Feature Elimination in Random Forest to Account for Correlated Variables in High Dimensional Data. *BMC Genet.* **2018**, *19* (S1), 65. <https://doi.org/10.1186/s12863-018-0633-8>.
- (56) Ludwig, S. M.; Natali, S. M.; Mann, P. J.; Schade, J. D.; Holmes, R. M.; Powell, M.; Fiske, G.; Commene, R. Using Machine Learning to Predict Inland Aquatic CO₂ and CH₄ Concentrations and the Effects of Wildfires in the Yukon-Kuskokwim Delta, Alaska. *Glob. Biogeochem. Cycles* **2022**, *36* (4). <https://doi.org/10.1029/2021GB007146>.
- (57) Amos, H. M.; Jacob, D. J.; Holmes, C. D.; Fisher, J. A.; Wang, Q.; Yantosca, R. M.; Corbitt, E. S.; Galarneau, E.; Rutter, A. P.; Gustin, M. S.; Steffen, A.; Schauer, J. J.; Graydon, J. A.; Louis, V. L. St.; Talbot, R. W.; Edgerton, E. S.; Zhang, Y.; Sunderland, E. M. Gas-Particle Partitioning of Atmospheric Hg(II) and Its Effect on Global Mercury Deposition. *Atmospheric Chem. Phys.* **2012**, *12* (1), 591–603. <https://doi.org/10.5194/acp-12-591-2012>.
- (58) Rienecker, M. M.; Suarez, M. J.; Todling, R.; Bacmeister, J.; Takacs, L.; Liu, H.-C.; Gu, W.; Sienkiewicz, M.; Koster, R. D.; Gelaro, R.; Stajner, I.; Nielsen, J. E. *The GEOS-5 Data Assimilation System— Documentation of Versions 5.0.1, 5.1.0, and 5.2.0*; NASA/TM–2008–104606, Vol. 27; National Aeronautics and Space Administration, 2008. <https://ntrs.nasa.gov/citations/20120011955>.
- (59) Streets, D. G.; Horowitz, H. M.; Lu, Z.; Levin, L.; Thackray, C. P.; Sunderland, E. M. Global and Regional Trends in Mercury Emissions and Concentrations, 2010–2015. *Atmos. Environ.* **2019**, *201*, 417–427. <https://doi.org/10.1016/j.atmosenv.2018.12.031>.
- (60) Val Martin, M.; Logan, J. A.; Kahn, R. A.; Leung, F.-Y.; Nelson, D. L.; Diner, D. J. Smoke Injection Heights from Fires in North America: Analysis of 5 Years of Satellite Observations. *Atmospheric Chem. Phys.* **2010**, *10* (4), 1491–1510. <https://doi.org/10.5194/acp-10-1491-2010>.
- (61) Hugelius, G.; Loisel, J.; Chadburn, S.; Jackson, R. B.; Jones, M.; MacDonald, G.; Marushchak, M.; Olefeldt, D.; Packalen, M.; Siewert, M. B.; Treat, C.; Turetsky, M.; Voigt, C.; Yu, Z. Large Stocks of Peatland Carbon and Nitrogen Are Vulnerable to Permafrost Thaw. *Proc. Natl. Acad. Sci.* **2020**, 201916387. <https://doi.org/10.1073/pnas.1916387117>.
- (62) Gorelick, N.; Hancher, M.; Dixon, M.; Ilyushchenko, S.; Thau, D.; Moore, R. Google Earth Engine: Planetary-Scale Geospatial Analysis for Everyone. *Remote Sens. Environ.* **2017**, *202*, 18–27. <https://doi.org/10.1016/j.rse.2017.06.031>.
- (63) R Core Team. R: A Language and Environment for Statistical Computing, 2022.
- (64) Osterwalder, S.; Bishop, K.; Alewell, C.; Fritsche, J.; Laudon, H.; Åkerblom, S.; Nilsson, M. B. Mercury Evasion from a Boreal Peatland Shortens the Timeline for Recovery from Legacy Pollution. *Sci. Rep.* **2017**, *7* (1), 16022. <https://doi.org/10.1038/s41598-017-16141-7>.
- (65) Fahnestock, M. F.; Bryce, J. G.; McCalley, C. K.; Montesdeoca, M.; Bai, S.; Li, Y.; Driscoll, C. T.; Crill, P. M.; Rich, V. I.; Varner, R. K. Mercury Reallocation in Thawing Subarctic Peatlands. *Geochem. Perspect. Lett.* **2019**, 33–38. <https://doi.org/10.7185/geochemlet.1922>.
- (66) Kuhn, M. Building Predictive Models in R Using the **Caret** Package. *J. Stat. Softw.* **2008**, *28* (5). <https://doi.org/10.18637/jss.v028.i05>.
- (67) Greener, J. G.; Kandathil, S. M.; Moffat, L.; Jones, D. T. A Guide to Machine Learning for Biologists. *Nat. Rev. Mol. Cell Biol.* **2022**, *23* (1), 40–55. <https://doi.org/10.1038/s41580-021-00407-0>.

- (68) Davies, G. M.; Gray, A.; Rein, G.; Legg, C. J. Peat Consumption and Carbon Loss Due to Smouldering Wildfire in a Temperate Peatland. *For. Ecol. Manag.* **2013**, *308*, 169–177. <https://doi.org/10.1016/j.foreco.2013.07.051>.
- (69) Hokanson, K. J.; Lukenbach, M. C.; Devito, K. J.; Kettridge, N.; Petrone, R. M.; Waddington, J. M. Groundwater Connectivity Controls Peat Burn Severity in the Boreal Plains. *Ecohydrology* **2016**, *9* (4), 574–584. <https://doi.org/10.1002/eco.1657>.
- (70) Lukenbach, M. C.; Hokanson, K. J.; Moore, P. A.; Devito, K. J.; Kettridge, N.; Thompson, D. K.; Wotton, B. M.; Petrone, R. M.; Waddington, J. M. Hydrological Controls on Deep Burning in a Northern Forested Peatland. *Hydrol. Process.* **2015**, *29* (18), 4114–4124. <https://doi.org/10.1002/hyp.10440>.
- (71) Santoso, M. A.; Huang, X.; Prat-Guitart, N.; Christensen, E.; Hu, Y.; Rein, G. Smouldering Fires and Soils. In *Fire Effects on Soil Properties*; Pereira, P., Mataix-Solera, J., Úbeda, X., Rein, G., Cerdà, A., Eds.; CSIRO Publishing, 2019; pp 203–216.
- (72) Turetsky, M. R.; Kane, E. S.; Harden, J. W.; Ottmar, R. D.; Manies, K. L.; Hoy, E.; Kasischke, E. S. Recent Acceleration of Biomass Burning and Carbon Losses in Alaskan Forests and Peatlands. *Nat. Geosci.* **2011**, *4* (1), 27–31. <https://doi.org/10.1038/ngeo1027>.
- (73) Walker, X. J.; Baltzer, J. L.; Bourgeau-Chavez, L. L.; Day, N. J.; De groot, W. J.; Dieleman, C.; Hoy, E. E.; Johnstone, J. F.; Kane, E. S.; Parisien, M. A.; Potter, S.; Rogers, B. M.; Turetsky, M. R.; Veraverbeke, S.; Whitman, E.; Mack, M. C. Arctic-Boreal Vulnerability Experiment (ABOVE) Synthesis of Burned and Unburned Forest Site Data, AK and Canada, 1983-2016, 2020, 0 MB. <https://doi.org/10.3334/ORNLDAAAC/1744>.
- (74) Wilkinson, S. L.; Moore, P. A.; Flannigan, M. D.; Wotton, B. M.; Waddington, J. M. Did Enhanced Afforestation Cause High Severity Peat Burn in the Fort McMurray Horse River Wildfire? *Environ. Res. Lett.* **2018**, *13* (1), 014018. <https://doi.org/10.1088/1748-9326/aa136>.
- (75) Jones, B. M.; Kolden, C. A.; Jandt, R.; Abatzoglou, J. T.; Urban, F.; Arp, C. D. Fire Behavior, Weather, and Burn Severity of the 2007 Anaktuvuk River Tundra Fire, North Slope, Alaska. *Arct. Antarct. Alp. Res.* **2009**, *41* (3), 309–316. <https://doi.org/10.1657/1938-4246-41.3.309>.
- (76) Zolkos, S.; MacDonald, E.; Hung, J. K. Y.; Schade, J. D.; Ludwig, S.; Mann, P. J.; Treharne, R.; Natali, S. Physiographic Controls and Wildfire Effects on Aquatic Biogeochemistry in Tundra of the Yukon-Kuskokwim Delta, Alaska. *J. Geophys. Res. Biogeosciences* **2022**, *127* (8). <https://doi.org/10.1029/2022JG006891>.
- (77) Horwath Burnham, J.; Sletten, R. S. Spatial Distribution of Soil Organic Carbon in Northwest Greenland and Underestimates of High Arctic Carbon Stores. *Glob. Biogeochem. Cycles* **2010**, *24* (3), 2009GB003660. <https://doi.org/10.1029/2009GB003660>.
- (78) Wu, T.; Wang, D.; Mu, C.; Zhang, W.; Zhu, X.; Zhao, L.; Li, R.; Hu, G.; Zou, D.; Chen, J.; Wei, X.; Wen, A.; Shang, C.; La, Y.; Lou, P.; Ma, X.; Wu, X. Storage, Patterns, and Environmental Controls of Soil Organic Carbon Stocks in the Permafrost Regions of the Northern Hemisphere. *Sci. Total Environ.* **2022**, *828*, 154464. <https://doi.org/10.1016/j.scitotenv.2022.154464>.
- (79) Fraser, A.; Dastoor, A.; Ryjkov, A. How Important Is Biomass Burning in Canada to Mercury Contamination? *Atmospheric Chem. Phys.* **2018**, *18* (10), 7263–7286. <https://doi.org/10.5194/acp-18-7263-2018>.

- (80) Turetsky, M. R.; Benscoter, B.; Page, S.; Rein, G.; van der Werf, G. R.; Watts, A. Global Vulnerability of Peatlands to Fire and Carbon Loss. *Nat. Geosci.* **2015**, *8* (1), 11–14. <https://doi.org/10.1038/ngeo2325>.
- (81) MDN. *National Atmospheric Deposition Program Mercury Deposition Network Interactive Map*. <https://nadp.slh.wisc.edu/maps-data/mdn-interactive-map/> (accessed 2024-05-19).
- (82) Baillargeon, N.; Pold, G.; Natali, S. M.; Sistla, S. A. Lowland Tundra Plant Stoichiometry Is Somewhat Resilient Decades Following Fire despite Substantial and Sustained Shifts in Community Structure. *Arct. Antarct. Alp. Res.* **2022**, *54* (1), 525–536. <https://doi.org/10.1080/15230430.2022.2121246>.
- (83) Wilkinson, S. L.; Andersen, R.; Moore, P. A.; Davidson, S. J.; Granath, G.; Waddington, J. M. Wildfire and Degradation Accelerate Northern Peatland Carbon Release. *Nat. Clim. Change* **2023**, *13* (5), 456–461. <https://doi.org/10.1038/s41558-023-01657-w>.
- (84) Ingram, R. C.; Moore, P. A.; Wilkinson, S.; Petrone, R. M.; Waddington, J. M. Postfire Soil Carbon Accumulation Does Not Recover Boreal Peatland Combustion Loss in Some Hydrogeological Settings. *J. Geophys. Res. Biogeosciences* **2019**, *124* (4), 775–788. <https://doi.org/10.1029/2018JG004716>.
- (85) Ackley, C.; Tank, S. E.; Haynes, K. M.; Rezanezhad, F.; McCarter, C.; Quinton, W. L. Coupled Hydrological and Geochemical Impacts of Wildfire in Peatland-Dominated Regions of Discontinuous Permafrost. *Sci. Total Environ.* **2021**, *782*, 146841. <https://doi.org/10.1016/j.scitotenv.2021.146841>.
- (86) Kumar, A.; Wu, S. Mercury Pollution in the Arctic from Wildfires: Source Attribution for the 2000s. *Environ. Sci. Technol.* **2019**, *53* (19), 11269–11275. <https://doi.org/10.1021/acs.est.9b01773>.
- (87) Giglio, L.; Randerson, J. T.; Van Der Werf, G. R. Analysis of Daily, Monthly, and Annual Burned Area Using the Fourth-generation Global Fire Emissions Database (GFED4). *J. Geophys. Res. Biogeosciences* **2013**, *118* (1), 317–328. <https://doi.org/10.1002/jgrg.20042>.
- (88) Giglio, L.; Schroeder, W.; Justice, C. O. The Collection 6 MODIS Active Fire Detection Algorithm and Fire Products. *Remote Sens. Environ.* **2016**, *178*, 31–41. <https://doi.org/10.1016/j.rse.2016.02.054>.
- (89) Lizundia-Loiola, J.; Otón, G.; Ramo, R.; Chuvieco, E. A Spatio-Temporal Active-Fire Clustering Approach for Global Burned Area Mapping at 250 m from MODIS Data. *Remote Sens. Environ.* **2020**, *236*, 111493. <https://doi.org/10.1016/j.rse.2019.111493>.
- (90) Hope, E. S.; McKenney, D. W.; Johnston, L. M.; Johnston, J. M. A Cost-Benefit Analysis of WildFireSat, a Wildfire Monitoring Satellite Mission for Canada. *PLOS ONE* **2024**, *19* (5), e0302699. <https://doi.org/10.1371/journal.pone.0302699>.
- (91) Zolkos, S.; Geyman, B. M.; Potter, S.; Moubarak, M.; Rogers, B. M.; Baillargeon, N.; Dey, S.; Ludwig, S. M.; Melton, S.; Navarro-Pérez, E.; McElvein, A.; Balcom, P. H.; Natali, S. M.; Sistla, S.; Sunderland, E. M. Substantial Mercury Releases and Local Deposition from Permafrost Peatland Wildfires in Southwestern Alaska, 2024. <https://doi.org/10.7910/DVN/ZA5JBZ>.

Table 1. Wildfire Hg emissions (E) and fire-attributable deposition (D) over three study domains: Alaska, the Yukon-Kuskokwim Delta (YKD), and Izaviknek and Kingaglia Uplands (IKU). Baseline = total Hg deposition from all sources excluding the IKU fires from May 2015 to May 2016. TGM = total gaseous mercury. TPM = total particulate mercury. f_{TGM} = fraction total Hg as TGM. Units in kg, except for f_{TGM} . Scenarios of Hg emissions (e.g., “Central E”) and f_{TGM} (e.g., Low f_{TGM}) constrain wildfire-attributable Hg deposition across the range of Hg emissions estimated in this study (mean = 556 kg, 5th-95th percentile = 164-1138 kg) and published f_{TGM} (93.5-99.2%) measured from peat fire.¹⁹ Hg E and D speciation and D_{Total} were simulated using the GEOS-Chem global atmospheric Hg model.

Domain of deposition	Area [km ²]	Baseline [kg]	Scenario (E / f_{TGM})	f_{TGM} (0-1)	E _{total} [kg]	E _{TGM} [kg]	E _{TPM} [kg]	D _{Total} [kg]	D _{TGM} [kg]	D _{TPM} [kg]
Alaska	3807281	42732	(1) Central / Low	0.935	556	519.9	36.1	74.8	52.8	22.0
Alaska	3807281	42732	(2) Low / Low	0.935	164	153.3	10.7	22.1	15.6	6.5
Alaska	3807281	42732	(3) Low / High	0.992	164	162.7	1.3	17.3	16.5	0.8
Alaska	3807281	42732	(4) High / Low	0.935	1138	1064.0	74.0	153.2	108.1	45.1
Alaska	3807281	42732	(5) High / High	0.992	1138	1128.9	9.1	120.3	114.7	5.6
YKD	304250	3489	(1) Central / Low	0.935	556	519.9	36.1	46.3	29.0	17.3
YKD	304250	3489	(2) Low / Low	0.935	164	153.3	10.7	13.6	8.5	5.1
YKD	304250	3489	(3) Low / High	0.992	164	162.7	1.3	9.7	9.1	0.6
YKD	304250	3489	(4) High / Low	0.935	1138	1064.0	74.0	94.7	59.3	35.4
YKD	304250	3489	(5) High / High	0.992	1138	1128.9	9.1	67.3	62.9	4.4
IKU	2777	49	(1) Central / Low	0.935	556	519.9	36.1	8.6	5.3	3.3
IKU	2777	49	(2) Low / Low	0.935	164	153.3	10.7	2.6	1.6	1.0
IKU	2777	49	(3) Low / High	0.992	164	162.7	1.3	1.7	1.6	0.1
IKU	2777	49	(4) High / Low	0.935	1138	1064.0	74.0	17.5	10.8	6.7
IKU	2777	49	(5) High / High	0.992	1138	1128.9	9.1	12.2	11.4	0.8

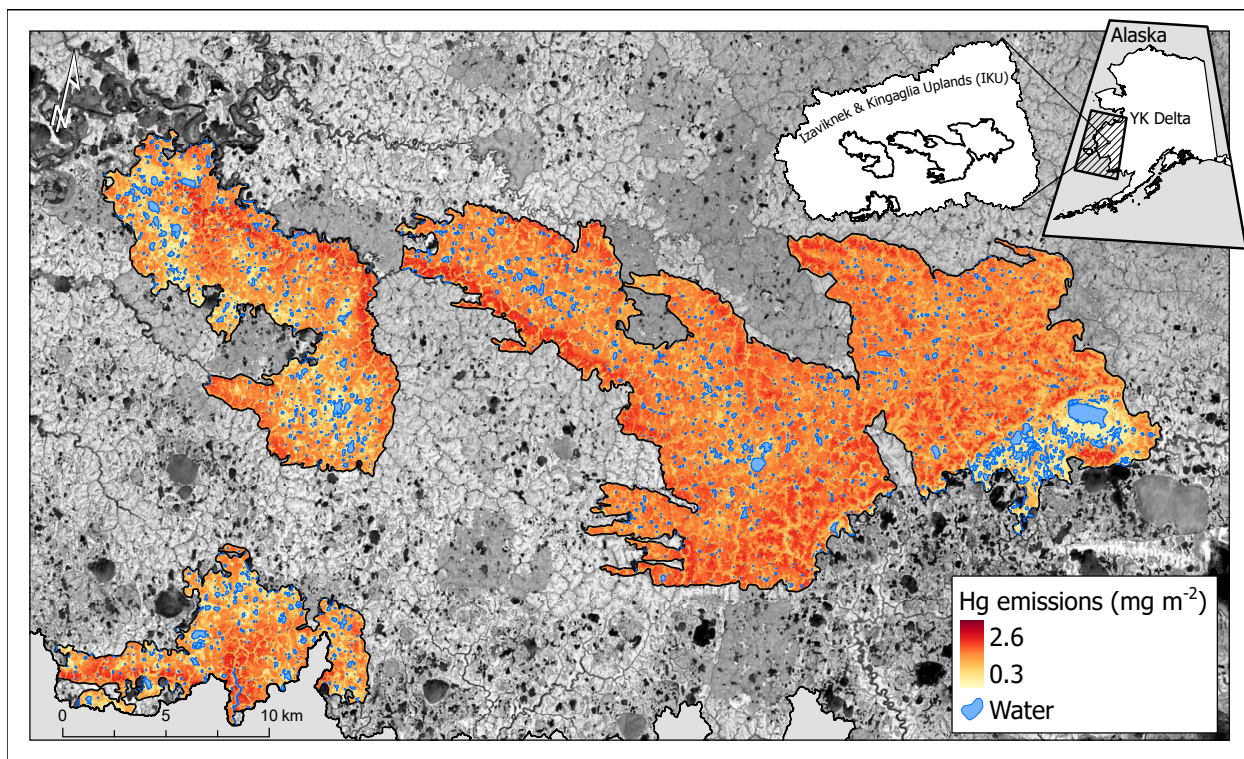


Figure 1. The distribution of terrestrial Hg release from wildfires during summer 2015 in the Izaviknek and Kingaglia Uplands (IKU) study area. Hg release was predicted using a machine learning (Random Forest) model, which was developed using measurements of soil and vegetation Hg stores, soil organic carbon stores, burn depth, and environmental indices derived from satellite remote sensing data. Inset maps show locations of wildfire perimeters in the IKU study area (black lines), and the extents for which atmospheric transport and deposition of wildfire Hg were modeled in Alaska (gray box), the Yukon-Kuskokwim Delta (hatched box), and the IKU. Base imagery obtained from Copernicus Sentinel-2a data (European Space Agency, <https://sentinel.esa.int/>, last access: 01 September 2023).

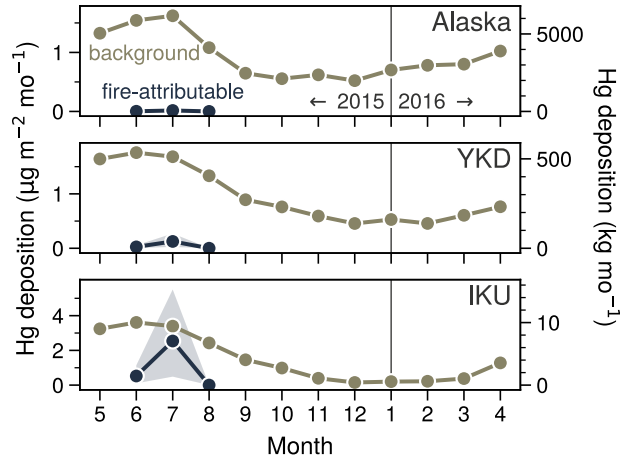


Figure 2. Total mercury (Hg) deposition from background sources (brown) and from the IKU fires (blue) over three domains: Alaska (top), the Yukon-Kuskokwim Delta (YKD; middle) and the Izaviknek and Kingaglia Uplands (IKU; bottom). Fire-attributable deposition is only shown for months where deposition occurred. The shaded blue area represents the range of uncertainty for fire-attributable deposition associated with uncertainties in plume injection altitude, Hg emission speciation, and Hg emission magnitude. The left axis shows areal deposition ($\mu\text{g m}^{-2} \text{mo}^{-1}$) and the right axis show cumulative deposition over each domain (kg mo^{-1}). Deposition was simulated using the GEOS-Chem global atmospheric Hg model.³⁸

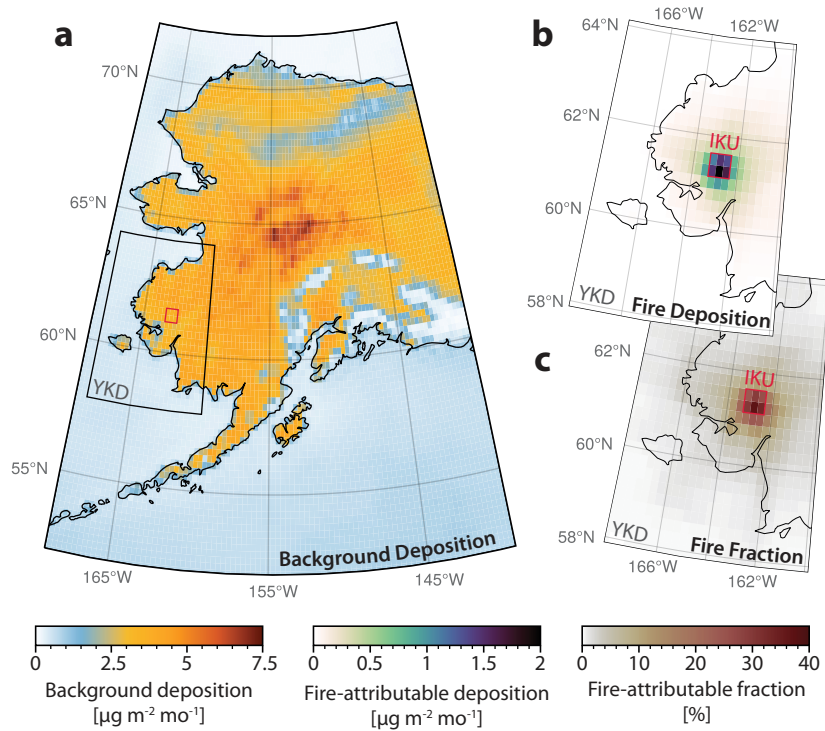


Figure 3. The distribution of Hg deposition during the period of active fires in the Izaviknek and Kingaglia Uplands (IKU; June – July, 2015) simulated using the GEOS-Chem global atmospheric Hg model. (a) Monthly mean Hg deposition from background sources and (b) from the IKU fires. (c) the IKU fire-attributable fraction of annual total Hg deposition (background and fire-attributable). YKD = Yukon-Kuskokwim Delta.

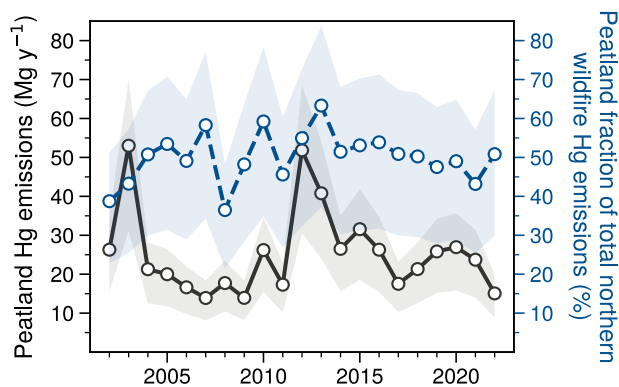


Figure 4. Annual peatland wildfire Hg emissions in the northern tundra-boreal region (>50°N) (black solid line) and the fraction of total annual wildfire Hg emissions in the northern tundra-boreal region that originate from peatlands (blue dashed line). Shaded regions represent the range of annual total and fractional peatland Hg emissions, estimated from the uncertainty in emissions from this study, as described in Sec. 2.5.3.