- 1 **Title:** Tracking disinfection by-products in northern Canada: impacts of dissolved organic matter
- 2 and regional variation
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- 16 Highlights
- Disinfection by-products were detected in all treated waters from eight northern communities
- of the Canadian Arctic, sometimes exceeding guidelines despite low dissolved organic matter
- 19 concentrations.
- Disinfection by-products formation was strongly linked to DOC levels, with additional
- 21 influences from pH and specific DOM fluorescent components.
- Climate-driven browning will likely increase DBP risks in northern communities, stressing the
- 23 need for better monitoring, treatment, and long-term adaptation.
- 24 Key words: Drinking water security, trihalomethanes, haloacetic acids, arctic lakes, water
- 25 treatment, chlorination, dissolved organic matter.
- 26 Data availability
- Data are available in Borealis repository: https://doi.org/10.5683/SP3/TUPBD3
- 28 To reviewers: please refer to the URL in the cover letter, as the data will be released only after the
- 29 paper is accepted.

#### Abstract

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31 The concentration of dissolved organic matter (DOM) is increasing in surface waters globally—a 32 phenomenon known as water browning—that is especially pronounced in northern ecosystems. 33 Browning affects lake mixing, light penetration, oxygen availability, and microbial communities, 34 and also presents new challenges for drinking water safety. In northern communities, where 35 drinking water is often treated without prior DOM removal, chlorination can lead to the formation 36 of disinfection by-products (DBPs), such as trihalomethanes (THMs) and haloacetic acids (HAAs), 37 both regulated in various countries for their potential health risks. In this study, we assessed DBP 38 concentrations in drinking water from eight northern communities located across the Northwest 39 Territories, Nunavut, and Nunavik, and examined their relationship with DOM quantity and 40 composition. Despite the oligotrophic nature of the source waters and their relatively low levels of 41 dissolved organic carbon (DOC < 6 mg L<sup>-1</sup>), DBPs were detected in all treated water samples, with 42 concentrations occasionally exceeding Canadian guidelines (up to 109 and 100 µg L<sup>-1</sup>, respectively). DBP formation was primarily correlated with DOC levels in source water and, to a 43 44 lesser extent, with pH and specific DOM components. Regional variations in DOM quality— 45 characterized using optical and fluorescence-based indicators—were also linked to differences in 46 DBP composition. These findings underscore the vulnerability of northern drinking water systems 47 to DBP formation, even under low-DOM conditions, and the importance of DOM quality as a 48 driver of DBP variability. As climate change intensifies browning through increased precipitation, permafrost thaw, and runoff, the associated risks of DBP formation in remote northern 49 50 communities are likely to rise, highlighting the need for enhanced monitoring, improved treatment 51 strategies, and long-term planning.

#### 1. Introduction

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53 Arctic ecosystems are responding to climate warming with evident features like coastal erosion 54 and landslides, and browning waters. Subtle changes are also expected in freshwater systems due 55 to shorter ice-cover periods, shifts in catchment vegetation, and increased nutrient inputs, all of 56 which impact the productivity and functioning of lakes and rivers (Calizza et al., 2022; Myers-57 Smith et al., 2011; Vonk et al., 2015). This function is closely tied to the ecological services 58 provided by freshwater ecosystems, including the supply of high-quality drinking water. 59 Dissolved organic matter (DOM) is an essential component of lake ecosystems as it controls 60 surface temperature and stratification strength, light and oxygen availability, and it acts as a source 61 of essential nutrients and carbon to microbial food webs (Creed et al., 2018). DOM is a complex 62 assemblage of molecules with varying origins and chemical properties that influence their 63 biological and photochemical reactivity (Battin et al. 2009). The concentration and chemical 64 properties of DOM are also crucial determinants of source water quality, as organic molecules 65 serve as precursors for the formation of disinfection by-products (DBPs) during drinking water 66 production (Zhang et al., 2018). 67 Lakes and rivers serve as vital sources of drinking water for communities that rely on the land for 68 their traditional activities (Martin et al., 2007). As these communities transition to settled hamlets, the use of more localized water sources has necessitated water treatment (Daley et al., 2015). Small 69 70 communities living in remote areas face challenges related to the potential presence of harmful 71 microorganisms and contaminants, as treatment plants often rely solely on ultraviolet radiation and 72 chlorination to produce drinking water (Daley et al., 2018; Sarkar et al., 2015). These communities 73 frequently encounter boiling advisories (Moghaddam-Ghadimi et al., 2023). However, drinking 74 water quality, preferences, and supply have been little studied in Arctic communities (Cassivi et 75 al., 2023; Gora et al., 2020; Medeiros et al., 2017). 76 Climate change increases the vulnerability of drinking water sources to contamination and 77 shortages (Leveque et al., 2021). The rising concentrations of DOM in areas impacted by 78 permafrost thawing and erosion (Wauthy et al., 2018) are likely to affect drinking water quality in 79 northern communities, particularly in relation to microbial activity and the formation of 80 disinfection by-products (DBPs). For example, Daley et al. (2018) observed low microbial risks in

Arctic drinking water; however they noted the potential for bacterial regrowth and biofilm

- 82 formation in truck water distribution systems, which will be influenced by carbon inputs and
- warmer temperatures. Relatively high DBP concentrations were found in small systems supplied
- 84 by surface waters in Quebec, Newfoundland and Labrador, fluctuating seasonally (Guilherme et
- 85 al., 2014).
- 86 In this study, we analyzed the occurrence and composition of DBPs in drinking water from eight
- 87 northern Canadian communities and examined their relationship with DOM properties. This
- 88 research was conducted alongside an analysis of microbial communities throughout the water
- treatment process (Moisan et al., 2025). The goal was to enhance our understanding of future risks
- associated with permafrost thaw and water browning, ultimately supporting the adaptation of water
- 91 treatment strategies in these vulnerable regions.

### 92 **2. Methods**

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# 2.1. Sampling locations and dates

- Water samples were collected from eight communities between 2019 and 2021 (Fig. 1 and Table
- 95 S1). While the pandemic posed significant challenges, the collaboration with many northern
- partners enabled to achieve most of the project objectives. In summer 2019, water samples were
- 97 collected from Kangiqsualujjuaq (KAN), and Salluit (SAL) in Nunavik, Taloyoak (TAL), Pond
- 98 Inlet-Mittimatalik (MIT), and Baker Lake-Qamani'tuaq (QAM) in Nunavut, and Fort Good Hope
- 99 (FGH) in Northwest Territories. Despite the complete closure of access to the North in 2020, we
- were able to obtain water samples from FGH partners. In 2021, water samples were collected from
- 101 Cambridge Bay-Iqaluktuuttiaq (CB) in Nunavut and Kangirsuk (KGS) in Nunavik, along with
- additional samples from MIT, KAN and FGH.

#### 103 2.2. Water collection from source to tap

- Samples were collected at multiple stages of the water treatment process: (1) directly from the
- source water (SW; not available at SAL), (2) from raw water in the reservoir of the treatment plant
- (RW), (3) after the final treatment step in chlorinated water (CW), (4) from water in distribution
- trucks (TW; not available at KGS), (5) from household tap water (HO), and (6) from public taps
- 108 (PT; not available at TAL). Public taps are located at the treatment plant and dispense water that
- has been further filtrated (activated carbon or fine filtration) to provide better-tasting treated water.
- RW was used as a surrogate for SW when the latter was not sampled as is the case for communities
- of SAL and KAN. Table S2 summarizes the sample collection details and the water treatment

processes used in the various communities. The collected water samples were stored in coolers and shipped via fast courier in 2L polycarbonate bottles, and processed at the laboratory facilities of INRS within 4 days in most cases (up to 8 in 2021 for CB, FGH, and KAN). Only DBP samples were prepared *in situ* by partners (see below).



**Figure 1.** Sampling location of the eight hamlets from which water samples were collected: Fort Good Hope (FGH), Cambridge Bay (CB), Qamani'tuaq (Baker Lake, QAM), Taloyoak (TAL), Mittimatalik (Pond Inlet, MIT), Kangigsualujjuaq (Georges River, KAN), Kangirsuk (KGS), and Salluit (SAL).

#### 2.3. Phytoplankton biomass and total phosphorus

Water samples were prepared to quantify phytoplankton biomass (chlorophyll-a, Chl-a) and total phosphorus (TP) in order to assess the trophic status of the lakes. However, at FGH and SAL, the source water is a river rather than a lake (Table S2). To determine Chl-a concentration, water samples were filtered onto GF/F filters (0.3 μm nominal porosity; AMD Manufacturing) and stored at –80°C for subsequent pigment extraction using the warm ethanol technique (Nusch, 1980). For total phosphorus, water samples were stored in polypropylene tubes with H<sub>2</sub>SO<sub>4</sub> (final pH < 2), and concentration determined by colorimetry on an Astoria 2 analyzer (Astoria-Pacific, Clackamas, Oregon, USA) after the reduction of antimony-phospho-molybdate complexes by ascorbic acid (EPA Method 365.3), following a potassium persulfate digestion step (detection limit of 0.7 μg L<sup>-1</sup>).

# 2.4. Dissolved organic matter characterization

133 Dissolved organic carbon (DOC) concentrations were obtained by high-resolution mass 134 spectroscopy (Sievers M9 Portable TOC Analyzer) on previously filtered (GF/F, 0.3 µm nominal 135 porosity) and acidified samples (HCl, final pH below 2), while spectral absorbance and 136 fluorescence matrices were obtained on separate samples at natural pH. Spectral absorption scans 137 acquired by spectrophotometry (Cary300, Varian) allowed to characterize the chromophoric 138 fraction of DOM (CDOM). The absorption coefficient at 320 nm (a<sub>320</sub>, m<sup>-1</sup>) was used as a proxy 139 for CDOM concentration, the specific UV absorbance at 254 nm per unit DOC (SUVA, L mgC<sup>-1</sup> 140 m<sup>-1</sup>) as a proxy of DOM aromaticity, and the CDOM spectral slope calculated between 275 and 141 295 nm (S<sub>285</sub>) as a proxy for DOM molecular size. 142 Matrices of fluorescence were obtained by spectrofluorimetry (Cary Eclipse, Varian) to 143 characterize the fluorescent fraction of the DOM pool (FDOM). A Parallel Factor Analysis 144 (PARAFAC; Stedmon et al., 2003) was constructed with R software, package staRdom 145 (spectroscopic analysis of DOM in R, version 1.1.25). The model was developed initially using 146 121 fluorescence matrices collected from source to tap. Following the removal of outliers (41 147 matrices from noisy treated waters and samples with very high DOM), models with 4 to 6 148 components were validated by split-half analysis. The model with 6 compounds was found most 149 appropriate to describe the dataset since no recurrent pattern or very sharp peak was observed in 150 the residual of modeled samples, and the sum squared errors in excitation and emission were 151 significantly lower than for models with fewer components. Loadings of each component had 152 characteristics in lines with what should be expected of independent and non-interacting groups of 153 fluorophores in aqueous solution (Murphy et al., 2013). 154 The six-component model retained explains 99.4% of the fluorescence variance, with each 155 component expressed as the max fluorescence of the main peak (given in Raman units or RU) or 156 as a fraction (%) of the total fluorescence (F<sub>tot</sub>; the sum of the five components as a proxy for 157 FDOM). The model includes four humic-like terrestrial components (HT1-4), one humic-like 158 microbial component (HM1), and one protein-like component matching tryptophan type molecules 159 (Try; Fig. S1). Components were matched with the OpenFluor database (Murphy et al., 2014) using 160 a similarity index above 0.95. A description of each component is presented in Table S3.

# 2.5. Disinfection by-products and chlorine

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- DBPs were identified and quantified according to the Environmental Protection Agency (EPA)
- standards. Samples were collected in vials pre-treated with sodium thiosulfate to stabilize DBPs as
- some are volatile. Simultaneously, pH was measured with a pocket tester (pHTestr 5, Oakton), and
- 166 free and total chlorine concentrations were determined with a pocket colorimeter (DR300, Hach).
- 167 HAA and THM concentrations were analysed by gas chromatography (Andersson et al., 2019).
- Analyzed trihalomethanes (THMs) include 4 compounds: trichloromethane or chloroform (TCM),
- bromodichloromethane (BDCM), dibromo-chloromethane (DBCM), and tribromo-methane or
- bromoform (TBM). For THM analysis, 1 mL of sample and 1 mL of the internal standard at a
- 171 concentration of 20 ppb (EPA fortification solution at 2000 ppm, Supelco # cat. 47358-U) was
- transferred into a 20 mL chromatographic vial. THMs were extracted by solid-phase
- microextraction with a fiber (50/30 µm DVBC/CAR/PDMS, Supleco 57298-U) using an
- autosampler (RTC-PAL). Separation was performed by gas chromatography (GC TRACE 1300,
- Thermo Scientific) with an RXI-XLB column (30 m x 0.25 mm x 0.25 mm, Restek 13723, or
- equivalent) coupled to a single quadrupole mass spectrometry detector (ISQ LT, Thermo
- Scientific). The established limits of quantification (LOQs) were 0.8 µg L<sup>-1</sup> for TCM, 1.7 µg L<sup>-1</sup>
- for BDCM,  $1.5 \mu g L^{-1}$  for DBCM, and  $1.9 \mu g L^{-1}$  for TBM.
- 179 Analyzed haloacetic acids (HAA) include 6 compounds: monobromoacetic acid (MBAA),
- dibromoacetic acid (DBAA), monochloroacetic acid (MCAA), dichloroacetic acid (DCAA),
- bromochloroacetic acid (BCAA), and trichloroacetic acid (TCAA). Extractions were based on EPA
- Method 552.3 using liquid-liquid extraction with a methyl butyl ether solvent. Extracts were
- analyzed by gas chromatography as above, using a DB-1701 column (or equivalent) coupled to an
- electron capture detector (Perkin Elmer Autosystem XL with 63Ni radioactive source). The LOQs
- 185 are  $1.0 \mu g L^{-1}$  for each HAAs.

#### 2.6. Statistical analyses

- To assess the impact of treatment on Chl-a and TP concentrations, a one-way ANOVA was
- 188 performed, comparing treated versus untreated waters (data normalized using a +1 log
- transformation). A principal component analysis (PCA) was conducted to examine how DOM
- 190 quality variables (SUVA, S<sub>285</sub>, and the relative abundance of the six FDOM components) varied
- among hamlets. An additional PCA was performed specifically for the Nunavut dataset, which

exhibited distinct patterns in the initial PCA, to further explore regional variations as there were more dates and sites in Nunavut. Wilk's lambda test was used to evaluate observed differences. A two-way ANOVA was conducted on DOM variables to assess the effects of water treatment and site, and a posteriori Tukey's tests. An additional PCA was performed to investigate differences in DBP relative abundances among hamlets, similarly as it was done for DOM. A significance level of α = 0.05 was applied throughout. Canonical correlation analyses (CCAs) were then used to explore relationships between DBPs and DOM. The first CCA explored correlations between DOM quantitative variables (DOC, a320, F<sub>101</sub>),

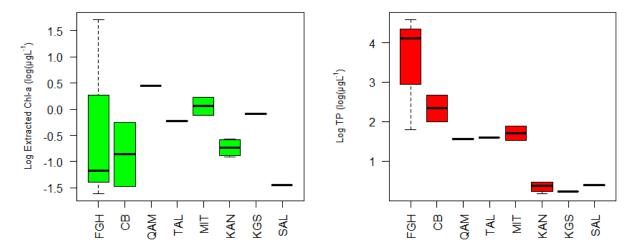
DOM. The first CCA explored correlations between DOM quantitative variables (DOC, a<sub>320</sub>, F<sub>tot</sub>), total chlorine (highly correlated with free chlorine; R=0.901, p<0.001), and pH with DBP concentrations (THMs and HAAs). The second CCA analysed the relationships between DOM quality variables (as defined above) and the relative abundance of DBPs (in % of total DBPs). The initial properties of DOM in raw waters (prior to UV treatment and chlorination) were linked to DBP production in truck waters (TW), which was collected from nearly all sampling sites. In KGS, only household tap water (HO) was available, as truck water was not sampled there. Pearson correlation tests were used to assess relationships between specific DOM variables and DBP components. Finally, to evaluate variations in DOM and DPB properties, a nested two-way ANOVA was conducted across different treatment stages. MCAA and TBM were excluded as their concentrations were below the LOQs in all samples, while BCAA, an unregulated HAA, was included since it exceeded the LOO.

#### 3. Results

#### 3.1. Biomass and nutrients in source and treated waters

Most municipal sources of drinking water studied had low concentrations of Chl-a and TP (Fig. 2; Table S4 presents selected variables by location, date, and water type; the complete dataset is available in the Borealis repository). The only exception was at FGH, where the source water from the Mackenzie River showed concentrations characteristic of mesotrophic to eutrophic systems. In this hamlet, however, treated water is drawn from a reservoir (RW) that is filled twice a year. For instance, the water treated in June 2019 came from the reservoir that had been filled in April of the same year. Overall, there was a reduction in Chl-a between source and treated waters (one-way ANOVA, p<0.001), and concentrations remained below 1 μg L<sup>-1</sup> after chlorination. Treated waters did not exhibit significantly lower phosphorus concentrations than source waters (one-way

ANOVA, p > 0.05), with the exception of FGH (1  $\mu$ g L<sup>-1</sup> on average; Fig. S2). Notably, a house tap at MIT, as well as the CW and TW at KAN, presented particularly high TP values.



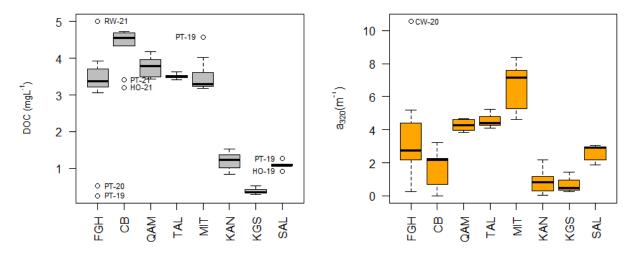
**Figure 2.** Concentrations of chlorophyll-a (Chl-a) and total phosphorus (TP) in source waters on a log scale. Whisker plots indicate minimum, maximum, quartiles (25, 50, 75, 100%), and median values for untreated waters (i.e. source waters) sampled on several dates (except for KGS, SAL, TAL and QAM that were only sampled once).

#### 3.2. DOM in source and treated waters

Source waters had DOC concentrations below 6 mg L<sup>-1</sup> and CDOM levels ( $a_{320}$ ) generally below 13 m<sup>-1</sup>, except at FGH ( $a_{320} = 33.7$  m<sup>-1</sup>). DOC was significantly lower in Nunavik waters ( $1.5 \pm 0.3$  mg L<sup>-1</sup>, n=30) compared to Nunavut ( $3.7 \pm 0.6$  mg L<sup>-1</sup>; n=39) or NWT waters ( $4.8 \pm 1.5$  mg L<sup>-1</sup>; n=17; p < 0.0001). However, this difference was not always reflected in CDOM values (Fig. S3 for source waters, Fig. 3 for treated waters). For instance, CB waters exhibited notably low CDOM relative to DOC concentration ( $a_{320} = 2.8$  m<sup>-1</sup> vs. an average of 7.5 m<sup>-1</sup> in other source waters, despite DOC in CB above 4 mg L<sup>-1</sup>), resulting in a particularly low SUVA index (Fig. 4). MIT waters had the highest CDOM and FDOM values, excluding FGH source water, which underwent significant reduction through settling in the reservoir (RW) and during coagulation/flocculation (Fig. 3, Table S4).

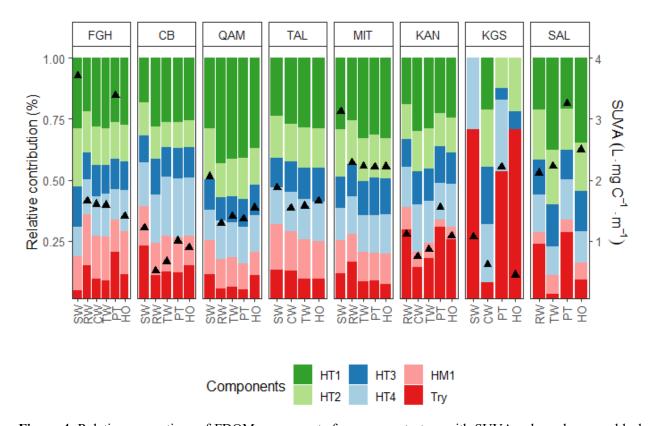
Most waters were dominated by humic-like terrestrial fluorophores, except in KGS, where fluorophores were dominated by tryptophan-like molecules (Try; Fig. 4). A PCA of DOM quality proxies further indicates that Nunavik waters had higher proportions of Try (Fig. 5A). In contrast, Nunavut waters were enriched in humic-like terrestrial fluorophores, with HT1 prevalent in QAM,

HT2-3 in MIT, and HT4 in CB (Fig. 5B). DOC concentration correlated most strongly to HT4 and HM1 (r > 0.71, p < 0.0001).

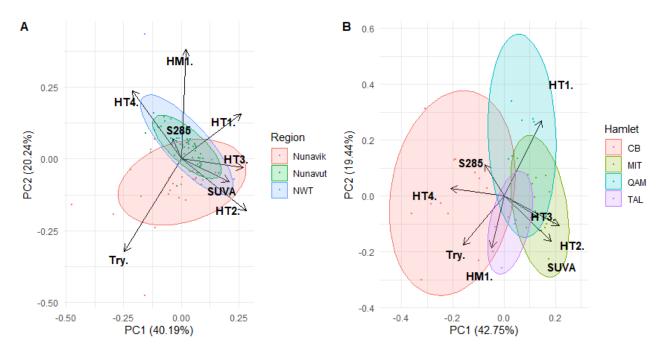


**Figure 3.** Concentrations of dissolved organic carbon (DOC) and colored dissolved organic matter (a<sub>320</sub>) in treated waters (raw reservoir water for FGH). Box-whisker plots indicate minimum, maximum, quartiles (25, 50, 75, 100%), and median values. Outliers are labeled with the corresponding water type name and year of sampling.

Water treatment (Table S2) did not significantly reduce DOC or CDOM in most communities, except at FGH, where treatment (settling in reservoir and coagulation/flocculation) resulted in reductions of 43% and 80%, respectively. DOC concentrations in FGH public taps were particularly low (<1 mg L<sup>-1</sup>; Fig. 3). Overall, water treatment caused minimal changes in DOM quantity and composition, with the only consistent and significant reduction observed in humic-like components at FGH (Table S5). The protein-like Try component showed slight decreases in most treated waters, but significant reductions occurred only in CB and KGS (p<0.01, Table S2). Notably, in KGS, the humic-like components HT2 and HT3 were detected only after chlorination, altering the relative abundance of fluorophores in tap waters (Fig. 4). The SUVA index decreased in treated water (p=0.02), with significant changes in FGH and QAM (p<0.03; Fig. 4).



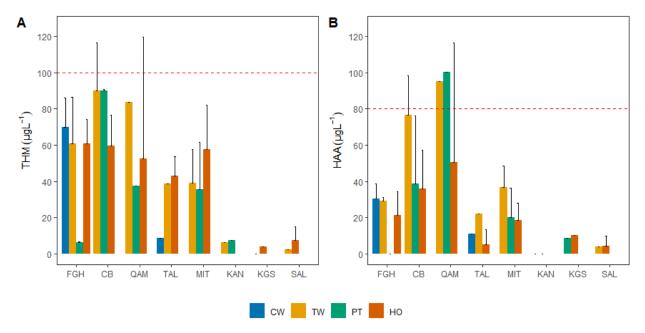
**Figure 4.** Relative proportions of FDOM components from source to tap, with SUVA values shown as black triangles. Components include four humic-like terrestrial compounds (HT1-4), one humic-like microbial compound (HM1), and one protein-like compound associated with tryptophan (Try).



**Figure 5.** PCA of DOM quality variables, including the six PARAFAC components (relative abundance), the SUVA index, and the spectral slope of DOM absorption at 285 nm ( $S_{285}$ ). (A) all hamlets; (B) Nunavut hamlets.

# 3.3. Disinfection by-products

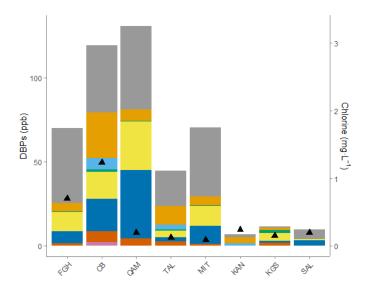
Chlorination in treatment plants, combined with sufficient DOM levels, led to the formation of DBPs in drinking water across all communities. On certain dates, DBP concentrations approached or exceeded Canadian guidelines for THMs (>100 µg L<sup>-1</sup>) and HAAs (>80 µg L<sup>-1</sup>) in FGH, QAM, CB, and MIT (Fig. 6). Notably, DBPs where above guidelines at CB on 26 July 2021 (TW), and at QAM on 18 Aug 2019 (TW, HO and PT), for DOC below 5 mg L<sup>-1</sup> and concentrations not particularly higher within the respective communities (Table S4). In contrast, DBP levels in Nunavik remained much lower, with THM below 12 µgL<sup>-1</sup> and HAA below 10 µgL<sup>-1</sup>. Similar to DOM patterns, DBP concentrations tended to be lower in Nunavik waters. A nested ANOVA, accounting for communities within regions, revealed significantly higher DBP concentrations in Nunavut, where QAM had the highest average concentration (131 µg L<sup>-1</sup>), followed by CB (119 µg L<sup>-1</sup>).



**Figure 6.** DBP concentrations measured along the water treatment process (CW: chlorinated water, TW: distribution truck water, PT: public tap, HO: house tap). (A) Sum of the four main trihalomethanes (THMs). (B) Sum of six haloacetic acids (BCAA included, since MCAA was below detection limit in all samples). Red dotted lines indicate the Canadian guideline values.

DBP composition varied among the communities, particularly in Nunavik waters where concentrations were much lower (Fig. 7 and 8). Chloroform (TCM) was the most abundant DBP in almost all sampled waters, accounting for 49% of total DBPs on average. However, in KGS (where DBP concentrations are among the lowest), it comprised only 6% of total DBPs, with a

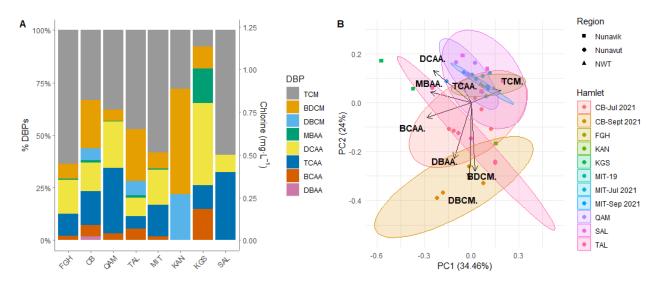
larger proportion of MBAA (17%) instead. This is the site where DOM had a particularly higher proportion of Try (protein-like fluorophores). DCAA and TCAA were the most abundant HAA measured, DCAA being particularly dominant in KGS but most abundant in QAM (Fig. 7). We did not detect any DBPs in the KAN samples from 2019 when the total chlorine concentration was notably low (<0.03 mg L<sup>-1</sup>). However, in 2021, low concentrations of THMs were detected (7 μg L<sup>-1</sup>) when the total chlorine concentration was higher (0.24 mg L<sup>-1</sup>). A PCAs on relative abundances of the different by-products (Fig. 8B; the first two PCA components explaining 58% of the variance) indicate notably the specific DBP composition of CB waters, particularly in September. The DBP composition in KAN and SAL treated waters was distinct and less diverse compared to other profiles, but these differences should be interpreted cautiously as quantities were sometimes approaching LOQs. We found a strong correlation between concentrations of TCM and TCAA among all treated waters (r=0.63 p<0.0001). A slight decrease in THM and HAA concentrations was observed in house tap water (HO) as compared to levels recorded in distribution trucks and chlorinated water, particularly for CB and QAM (Fig. 6), potentially related to the storage period of water in home tanks (some DBPs are volatile).



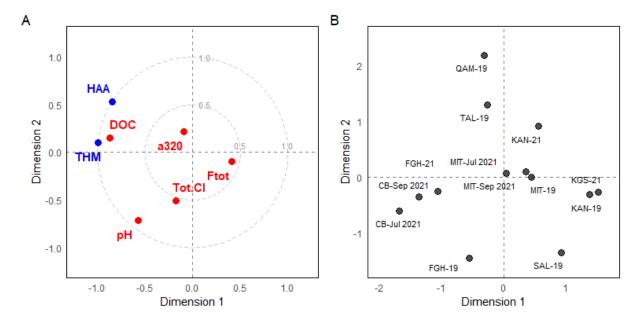
**Figure 7.** DBP concentrations in each hamlet, averaged across all treated water samples. Mean total chlorine concentrations are also shown as black triangles.

A canonical correlation analysis (CCA) was conducted to further assess the influence of source water DOM properties, total chlorine applied, and pH on the concentrations and composition of DBPs in truck waters (Fig. 9). The results are presented as biplots, illustrating the first two canonical dimensions. In Figure 9A, which links DOC concentration, a<sub>320</sub> (CDOM), and F<sub>tot</sub>

(FDOM) to total THMs and HAAs (individual quantities did not bring specific information), DOC emerged as the dominant variable on the first canonical dimension (r=-0.74), most closely associated with both THMs (r=-0.90) and HAAs (r=-0.80). The second dimension primarily captured the influence of pH (r=-0.80) and total chlorine (r=-0.50) on HAAs (r=0.29). The corresponding distribution plot (Fig. 9B) shows how specific water samples align along these patterns. For example, the elevated DOC levels in CB, and the low DOC levels in KAN and KGS were most strongly linked to differences in DBP concentrations. These associations are further supported by Pearson correlations showing strong positive correlations between DOC and both THMs (r=0.74, p=0.004) and HAAs (r=0.67; p=0.01). A second CCA (Fig. S4) explored how DOM quality variables—specifically the most relevant components selected by the model (HT2, HM1, Try, and SUVA)—relate to the relative abundance of individual DBP species (%TCM, %BDCM, %TCAA, and %DCAA retained). The first canonical dimension was primarily associated with HM1 (r=-0.85) and correlated most strongly to TCM and DCAA (r ranging from -0.34 to 0.25). The second dimension, associated to Try, HT2 and SUVA (r=-0.63, 0.86 and 0.74, respectively), related to all four DBPs but most strongly to BDCM (r=-0.57).



**Figure 8.** (A) Relative abundances (%) of DBPs averaged across treated waters. (B) PCA illustrating differences in DBP composition among hamlets and sampling years.



**Figure 9.** Canonical correlation analysis relating quantitative DOM properties (DOC, a<sub>320</sub>, F<sub>tot</sub>; in red), pH, and total chlorine (Tot.Cl) to DBP concentrations (sum of the four THMs and five HAAs; in blue), showing (A) the variables and (B) the truck water samples identified by hamlet and year (home tap water for KGS).

#### 4. Discussion

# 4.1. The trophic status of source waters and their organic matter content

As expected for water bodies used in drinking water production, the lakes studied in Nunavut and Nunavik can be classified as oligotrophic based on their Chl-a and TP concentrations (i.e., Chl-a  $< 3~\mu g~L^{-1}$  and TP  $< 10~\mu g~L^{-1}$ ). However, it is important to note that the trophic status of a lake is generally determined over more than one or two visits to a single station, ideally from at least two sampling stations, with monthly sampling over a period of three years (as per Schallenberg and van der Zon, 2021). Arctic lakes typically present low phytoplankton abundance (Ayala-Borda et al., 2021), which is strongly regulated by seasonality, low temperatures, and limited nutrients. In contrast, the Mackenzie River, the source water at FGH, presented much higher TP and Chl-a concentrations. As a dynamic lotic ecosystem, it transports suspended solids and nutrients from a vast catchment area (Emmerton et al., 2008). However, once stored in the reservoir–from which water is drawn for treatment–particle settling over several weeks is likely to substantially lower concentrations, as observed in 2021 (detailed results in Borealis repository). The extent of this reduction likely depends on the time elapsed since river pumping.

The source waters overall were also very transparent, with DOC concentration remaining below 6 mg L<sup>-1</sup> throughout the studied periods. Waters in Nunavik presented approximately 40% lower concentrations of DOC (and CDOM) compared to those in Nunavut, while water samples near FGH showed the highest concentrations and variability (Fig. S3). These variations are attributed to climatic and altitudinal differences between the regions, which influence vegetation patterns, permafrost coverage, and thermokarst activity (Osburn et al. 2017). To remove excess natural organic matter and limit the formation of DBPs, drinking water treatment plants typically employ a coagulation-flocculation steps (Matilainen et al., 2010). This process is used at the FGH treatment plant, where sedimentation in a reservoir is followed by coagulation-flocculation and filtration, with chlorination applied to clear water afterward (see section on water treatment effects below). Although the water bodies studied were primarily oligotrophic and transparent, these conditions may change in the future. Permafrost thawing could increase nutrient levels during the summer, promoting primary production and organic matter cycling (Creed et al., 2018). Additionally, the supply of nutrients-potentially amplified by intensified erosion from extreme precipitation eventscombined to the warmer waters, longer summers, and stronger stratification predicted for northern lakes, could stimulate cyanobacterial growth (Przytulska et al., 2017; Taranu et al., 2015). Given the potential for rapid changes as the climate evolves, it is crucial to enhance monitoring of the trophic status of source waters. Implementing simple, early-warning tools could provide valuable insights and enable timely interventions to safeguard water quality in these vulnerable ecosystems.

# 4.2. DOM characteristics in northern drinking waters

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The composition of DOM is known to influence DBP formation, particularly the presence of humic-like substances (Fernández-Pascual et al. 2023). Therefore, understanding the DOM composition in drinking water and its relationship with DBP formation could provide valuable insights for developing simple, accessible monitoring tools (e.g., pocket fluorimetry). In this study, humic-like molecules of terrestrial origin accounted for 72% of FDOM in the sampled waters, with clear regional variations in composition (Fig. 5). The relative importance of protein-like molecules (Try) was greater in Nunavik source waters (43% on average, as compared to 11% in Nunavut and FGH). Try was particularly important in source or raw waters of KGS (70%) and KAN (on avg 28%) (Fig. 4), potentially indicating larger inputs by primary producers or biofilms (Li et al., 2020), and limited microbial activity consuming these molecules. The presence of these biolabile

fluorophores in home taps suggests that they may support microbial activity in household reservoirs.

380 The relatively low SUVA values (generally < 3 L mgC<sup>-1</sup> m<sup>-1</sup>) suggest that aromatic molecules made 381 up less than 30% of the DOM pool (Weishaar et al., 2003), a pattern commonly observed in Arctic 382 lakes and rivers (Coch et al., 2019; Fowler et al., 2020). This implies that a significant portion of 383 the DOC in the studied waters was non-chromophoric and thus not effectively captured by the 384 characterisation methods employed here (fluorescent matrices). The SUVA index showed weak 385 correlations with any of the fluorophores, suggesting that aromatic molecules (with conjugated 386 C=C double bonds) were not dominant relative to other organic molecules (Hidayah et al., 2020). 387 Given this diversity of organic molecules, which likely varies by region and season, further 388 characterization methods that account for non-chromophoric molecules are needed (see the review 389 by Folhas et al., 2025). This will improve the understanding of how these compounds may interact 390 with chlorine to form DBPs. Nevertheless, certain trends are emerging from the analyses of CDOM 391 and FDOM (see below).

# 4.3. The effect of water treatment on water quality

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Water treatment resulted in a substantial reduction in Chl-a concentrations (Table S4), while storage in household reservoirs (HO compared to TW, or CW when TW was not available) did not lead to appreciable increases in phytoplankton biomass. The most notable reduction in TP between source and treated waters was observed at FGH, where coagulation and filtration are employed. These processes are well-known to effectively decrease nutrient levels and the abundance of phytoplankton (O'Melia, 1998). A few outliers in TP concentrations (treated waters in MIT and KAN; Fig. S2) may indicate contamination during sample handling, either at the sampling site or during laboratory processing (Holeton et al., 2011). Given the low phosphorus concentrations in drinking water, often approaching the methodological detection limit, extra precautions should be taken to prevent contamination.

Coagulation-flocculation is not employed in the treatment plants of Nunavut and Nunavik due to the relatively low levels of DOM in the source waters. This explains why none of the quantitative DOM indicators (DOC, a<sub>320</sub>, and F<sub>tot</sub>) showed significant changes between the source and treated waters (maximum a<sub>320</sub> reduction of only 7%). However, water treatment at FGH resulted in a substantial reduction of CDOM and SUVA (Table S4). Coagulation-flocculation is known to

408 effectively reduce DOM, particularly the hydrophobic fraction and high-molar-mass compounds 409 (Jiang, 2015; Matilainen et al., 2010). Among the fluorophore components, HT2 and HM1 were 410 most affected by treatment (HT2 reduced by 15-70%, HM1 by 6-57%). Studies have shown that 411 HT2 is sensitive to photodegradation (Zhou et al., 2019), making it susceptible to the UV treatment 412 applied. While HM1 was not a dominant FDOM component, its relative abundance showed a 413 strong correlation with DOC. This group of fluorophores, referred to as humic-like microbial 414 substances, serves as an indicator of recent biological sources of DOM (e.g. plant leachates; Pacoureau et al., 2025) and is often enriched in lakes with moderate to high primary production 415 416 (Osburn et al., 2017). In some cases, it has proven useful in tracking DOC concentrations 417 throughout water treatment (Shutova et al., 2014). Despite the large fraction of non-chromophoric 418 molecules in the studied waters, optimizing fluorimetric methods for detecting HM or HT 419 molecules could offer a valuable approach for monitoring DBP precursors in source waters used 420 for drinking.

# 4.4. Disinfection by-products in Arctic drinking water and their link to DOM

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Despite limited access to Arctic regions during the COVID-19 pandemic and restrictions on field sampling, our study revealed significant levels of DBPs in treated waters, particularly in Nunavut, where concentrations occasionally approached or exceeded regulatory guidelines (Fig. 6). Previous studies have demonstrated that DBP formation is influenced by both the concentration of DOM and chlorine dosage (Harir et al., 2019; Krasner et al., 2006; Sadiq et al., 2004). Although chlorine dosage in the treatment plants is not available, chlorine concentrations measured in treated waters showed limited variation across communities (total chlorine ranging from 0 to 0.66 mg L<sup>-1</sup>; Table S4), with most tap water samples containing concentrations below 0.1 mg L<sup>-1</sup>. These low concentrations suggest that chlorination may have been insufficient, but they lead to limited DBP formation, particularly in Nunavik communities. Moreover, the lower DBP concentrations observed in tap waters (compared to CW and TW) are likely attributable to the volatilization of THMs or the microbial degradation of HAAs (Rodriguez and Duchaine, 2013). Nevertheless, we found a strong positive correlation between DOC (in source waters) and total DBP concentrations (in TW; r=0.73; p=0.005), consistent with observations in UK water distribution systems (Bond et al. 2014). This suggests that even with standard chlorination protocols, elevated DOM levels particularly in the context of ongoing water browning in northern regions—can lead to increased DBP formation.

439 DBP production is commonly linked to the presence of aromatic and humic compounds in source 440 water (Fernández-Pascual et al., 2023; Hua et al., 2020). In our study, very low DBP levels in 441 Nunavik drinking waters likely reflected both lower chlorine application and reduced humic 442 content in raw waters. While no strong, consistent patterns emerged between overall DBP 443 composition or concentration and DOM characteristics (Fig. 9 & S4), we observed that atypical 444 DOM signatures—as seen in KGS—were associated with distinct DBP profiles, suggesting that a 445 broader and more systematic sampling effort could reveal stronger linkages. For example, CB and 446 SAL showed similar CDOM concentrations (Fig. 3 & S3) and comparable total residual chlorine 447 levels (0.36 mg L<sup>-1</sup> in CB; 0.53 mg L<sup>-1</sup> in SAL), yet DBP concentrations were considerably higher in CB (Fig. 6). In that hamlet, DOC was 78% higher and SUVA 34% lower than in SAL, suggesting 448 449 that non-colored DOM may also contribute to DBP formation. Further characterisation of this 450 uncolored DOM fraction is needed to clarify these dynamics and improve predictive capabilities. 451 Other influential water characteristics—such as temperature and pH, as well as the content of 452 inorganic ions such as bromide (impacting mostly DBP speciation)—are also known to influence 453 DBP formation (Zhang et al., 2020) and may have contributed to the observed patterns. Notably, 454 the elevated concentrations of MBAA and DBAA in CB could be associated with the presence of 455 bromide in Greiner Lake (not measured). 456 While SUVA is widely used as a proxy for aromatic DOM and a predictor of primary precursors 457 to THMs, HAAs, and other DBPs (Weishaar et al., 2003), our results did not reveal a significant 458 correlation between SUVA and DBP concentrations. This lack of association may be expected 459 given the diversity of drinking water sources and treatments applied, where factors such as 460 chlorination dose, water temperature, and pH can obscure such relationships. These findings align 461 with previous studies suggesting that SUVA may be more closely associated with non-regulated 462 or unidentified DBPs (Hua et al., 2015; 2020). However, the CCA linking DOM composition with 463 relative DBP composition showed that SUVA was associated with certain compounds, notably DCAA (Fig. S5). In waters with low SUVA values (< 2 L mg C<sup>-1</sup> m<sup>-1</sup>), DOM was shown to contain 464 465 hydrophilic components with amino and carboxyl groups, which are key precursors of DCAA and 466 brominated DBPs (Li et al., 2014). These DBPs were relatively abundant in our dataset, particularly 467 in CB, where they accounted for over 35% of total DBPs. Thus, while SUVA may not be a reliable 468 predictor of total DBP formation, specific DOM characteristics can help explain DBP profiles and 469 should be further investigated.

470 Among individual DBPs, TCM was the most prevalent across all sites, consistent with observations 471 in many other treated water systems (e.g., Rodriguez et al., 2007). TCM is known to originate from 472 various hydrophobic organic molecules, including humic substances (Hung et al., 2017; Li et al., 473 2014). Its relative abundance correlated strongly with that of the HT2 fluorophore (r=0.73, 474 p=0.005), which was consistently detected in source waters (Fig. 4), reinforcing the role of HT2 as 475 a key precursor of TCM. In KGS, water treatment significantly reduced the HT4 component, which 476 correlated with BDCM relative abundance (r=0.62, p=0.02). Fluorophores such as HT4 have 477 previously been identified as precursors effectively removed by magnetic ion exchange resins 478 (Jutaporn et al., 2022), supporting their potential as targets for improved DBP mitigation strategies.

# 5. Conclusions

- The water sources studied were generally of high quality for drinking purposes, characterized as oligotrophic and low in DOM during the sampled seasons. Despite overall low DOC levels (all below 6 mg L<sup>-1</sup>), significant regional differences in both the quantity and composition of DOM were observed. Disinfection by-products were nonetheless detected in all treated water samples, with some levels exceeding Canadian regulatory guidelines. DBP levels were particularly low in Nunavik, where chlorination doses (secondary disinfection) appeared insufficient to maintain adequate levels of residual chlorine concentrations.
- DOC emerged as the strongest predictor of total DBP concentrations, whereas other indicators, such as SUVA and FDOM, showed inconsistent relationships with DBP levels. This variability is likely due to the wide range of source water characteristics and treatment conditions, including differences in chlorine dose, water temperature, pH, and the presence of bromide. Although FDOM was dominated by humic-like molecules, SUVA values indicated relatively low aromaticity, implying the presence of a substantial fraction of uncolored and hydrophilic DOM–an under-characterized pool that may contribute significantly to DBP formation.
- With the anticipated browning of northern lakes and rivers—driven by longer, warmer summers, increased vegetation productivity, greater precipitation, and permafrost thaw—the risk of DBP formation, including regulated compounds like chloroform, is expected to increase. In our study, chloroform was the most prevalent DBP, and its relative abundance showed a moderate correlation with humic-like fluorophores, indicating a potential link to specific DOM components.

- 499 Furthermore, the application of more standard chlorination practices in treatment plants may
- 500 exacerbate this trend.
- These findings underscore the importance of enhanced DOM monitoring in northern source waters,
- as a key precursor for DBP formation. Establishing early-warning systems based on DOM
- 503 characteristics could improve risk management and support pro-active water treatment strategies.
- 504 Ultimately, incorporating coagulation-flocculation steps into treatment processes, as demonstrated
- in FGH, may offer an effective approach to reduce DOM content prior to chlorination and thereby
- 506 mitigate DBP formation, helping to ensure the long-term safety of drinking water in northern
- 507 communities.

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#### 515 Author contributions

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- 517 JC, IL; Methodology: CH, IL; Project administration: JC, IL; Supervision: IL; Writing original
- 518 draft: CH, IL; Writing review & editing: IL, CH, with a contribution of all co-authors.

#### 519 Data availability

- Data are available in Borealis repository: https://doi.org/10.5683/SP3/TUPBD3
- To reviewers: please refer to the URL in the cover letter, as the data will be released only after the
- 522 paper is accepted.

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# 679 Supplementary tables

# **Table S1.** Dates of the sampling campaigns in each hamlet.

Region	Hamlet	Summer 2019	Summer 2020	Spring/Summer 2021	Fall 2021
Northwest Territories	Fort Good Hope (FGH)	17 June	28 July	27 July	
Nunavut	Cambridge Bay (CB) Qamani'tuaq (QAM)	18 August		26 July	29 Sept
	Taloyoak (TAL) Mittimatalik (MIT)	29 July 20 June		29 April +17 August	
Nunavik	Salluit (SAL) Kangirsuk (KGS)	27 August		16 August	
	Kangiqsualujjuaq (KAN)	1 Sept		28 March+ 29 June	7 Sept

**Table S2.** Names of municipal source waters (SW), along with the sampling locations across the treatment process for each hamlet: raw water (RW), chlorinated water (CW), distribution truck water (TW), household tap (HO), and public tap (PT). The treatments applied at the treatment plant in each hamlet are also provided.

Hamlet	Source water	Collected samples	Treatment applied at 1866 treatment plant		
Fort Good Hope (FGH)	Mackenzie River	SW, RW, CW, TW, PT, HO	Sedimentation (reserved),7 coagulation, membrane 8 filtration, chlorination		
Cambridge Bay (CB)	Greiner Lake	SW, RW, TW, PT, HO	Membrane filtration, 689 chlorination 690		
Qamani'tuaq (QAM)	Baker Lake	SW, RW, TW, PT, HO	Chlorination 691		
Taloyoak (TAL)	Canso Lake	SW, RW, CW, TW, HO	UV, chlorination 692		
Mittimatalik (MIT)	Water Lake	SW, RW, TW, PT, HO	Chlorination 693 694		
Salluit (SAL)	Kuuguluk River	RW, TW, PT, HO	UV, chlorination 695		
Kangirsuk (KGS)	Tasialuk Lake	SW, CW, PT, HO	UV, chlorination 696		
Kangiqsualujjuaq (KAN)	Not named lake	RW, TW, CW, PT, HO	UV, chlorination 697		

**Table S3.** Characteristics of the six fluorophore components extracted by PARAFAC analysis on dissolved organic matter.

Component	Excitation (nm)						
HT1	255 / 325	440	Humic-like terrestrial, with high molecular weight and	matches 41			
			aromatic fluorescence				
HT2	275	520	Humic-like terrestrial, correlated to lignin phenols, parented	53			
			to fulvic acids				
HT3	280 / 370	458	Humic-like terrestrial, from lignin and other terrestrial	13			
			plant-derived precursor materials				
HT4	260	458	Humic-like terrestrial, indicative of DOM transformation	30			
			and degradation within lakes				
HM1	310	384	Humic-like, microbially processed autochthonous	43			
			compounds, linked to the degradation of both				
			algal and terrestrial sources				
Try	280	332	Tryptophan-like, microbial-delivered autochthonous,	125			
			correlated to terrestrial fluorophores				

**Table S4.** Selected variables in source waters (SW) and treated waters (truck water, TW) by location and date, including phytoplankton biomass (Chl-a), total phosphorus (TP), dissolved organic carbon (DOC), colored dissolved organic matter (a<sub>320</sub>), SUVA index, pH, total chlorine, trihalomethanes (THMs), and haloacetic acids (HAAs). The full dataset is available in the Borealis data repository.

Date	Water	Chl-a	TP	DOC	a <sub>320</sub>	SUVA	pН	Chlorine	THMs	HAAs		
Year	type	μg L <sup>-1</sup>	mg L <sup>-1</sup>	mg L <sup>-1</sup>	$m^{-1}$	L mgC <sup>-1</sup> m <sup>-1</sup>		mg L <sup>-1</sup>	μg L <sup>-1</sup>	μg L <sup>-1</sup>		
Fort Good Hope FGH												
17 June	$SW^1$	0.31	97.9	5.3	11.4	2.7						
2019	TW	0.02	1.7	3.4	5.1	2.0	8.6	0.49	79	28		
27 July	$SW^1$	5.53	61.0	5.6	12.6	2.8						
2021	$HO^2$	0.07	3.1	3.8	2.8	1.4	8.4	0.34	52	31		
Cambridge Bay CB												
26 July	SW	0.23	14.6	4.3	2.9	1.2						
2021	TW	0.02	8.9	4.7	0.1	0.5	8.4	0.34	109	92		
29 Sept	sw	0.78	7.4	4.5	2.8	1.2						
2021	TW	0.02	9.8	4.7	2.3	0.9	8.2	0.38	72	62		
	Qan	nani'tuaq	QAM									
18 Aug	sw	1.57	4.8	3.8	6.8	2.1						
2019	TW	0.04	8.3	4.0	4.6	1.4	6.4*	0.04	84	95		
	Talo	yoak TA	L									
29 July	sw	0.79	4.9	3.8	5.3	1.9						
2019	TW	0.05	6.1	3.5	4.3	1.6	7.6	0.21	39	22		
	Mitt	timatalik	MIT									
20 June	sw	0.89	4.6	3.9	7.5	2.3						
2019	TW	0.29	12.9	3.6	6.9	2.2	7.9	0.10	56	44		
17 Aug	sw	1.26	6.6	2.1	6.4	3.9						
2021	TW	0.15	4.9	3.3	4.6	1.9	7.6	0.66	42	43		
	Sall	uit SAL										
27 Aug	$RW^3$	0.23	1.5	1.1	1.8	2.1						
2019	TW	0.02	1.9	1.1	2.2	2.2	7.1	0.53	2	4		
	Kangirsuk KGS											
16 Aug	sw	0.91	1.3	0.4	1.0	1.1						
2020	$HO^2$	0.01	1.9	0.5	0.2	0.4	7.0	0.31	4	10		
	Kangiqsualujjuaq KAN											
1 Sept	$RW^3$	0.55	1.6	1.0	0.9	1.3						
2019	TW	0.23	1.7	1.3	0.4	0.6	6.8	0.01	0	0		
7 Sept	$RW^3$	0.40	1.2	1.6	0.7	0.7						
2021	TW	0.01	9.8	1.4	0.03	0.4	6.6	0.24	6	0		

<sup>&</sup>lt;sup>1</sup>Source waters were collected on the indicated sampling date, but the treated waters sampled on that date originated from the town reservoir, which had been filled at an earlier time (e.g., in April for the June 2019 sampling).

<sup>&</sup>lt;sup>2</sup>Values from home taps is reported in cases where truck waters data were incomplete.

<sup>&</sup>lt;sup>3</sup>Raw water (reservoir at the treatment plant) is reported in cases where source waters were not sampled.

<sup>\*</sup>Only measured in public tap for this hamlet.

# **Supplementary figures**

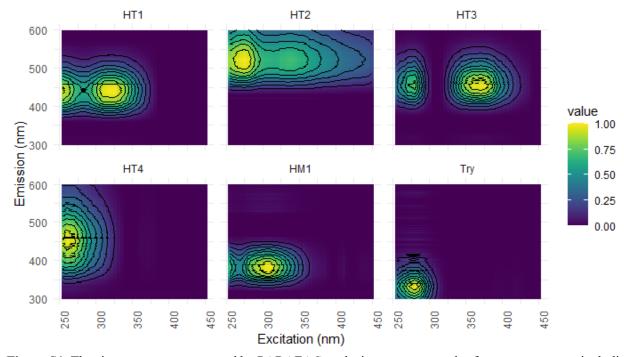
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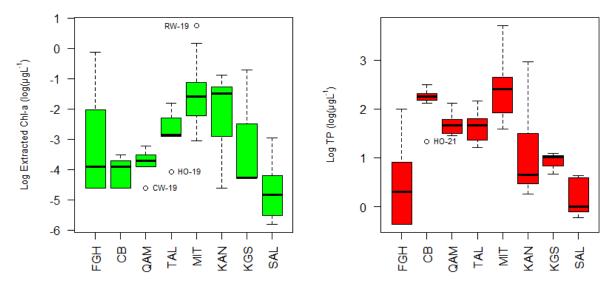
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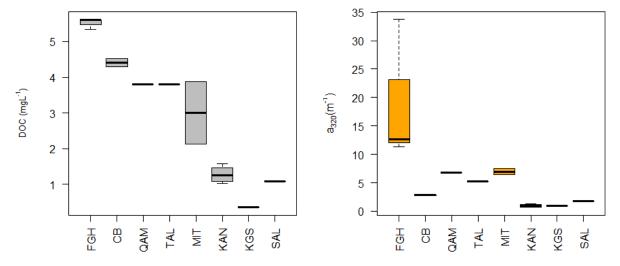
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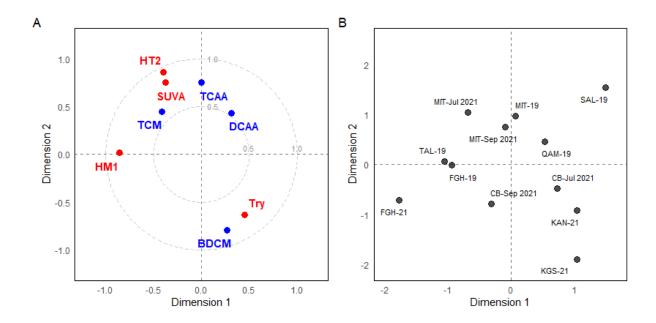
**Figure S1.** The six components extracted by PARAFAC analysis on water samples from source to tap, including four humic-like terrestrial components (HT1-4), one humic-like microbial component (HM1), and one protein-like components (tryptophan-like, Try).



**Figure S2.** Chlorophyll-a (Chl-a) and total phosphorus (TP) concentrations in treated waters on a log scale. Whisker-plots indicate minimum, maximum, quartiles (25, 50, 75, 100%), and median values for treated waters sampled on several dates (except for KGS, SAL, TAL and QAM only sampled once). Outliers are identified with their corresponding water types and year sampled.



**Figure S3.** Concentrations of dissolved organic carbon (DOC) and colored dissolved organic matter (a<sub>320</sub>) in source waters. Box-whisker plots indicate minimum, maximum, quartiles (25, 50, 75, 100%), and median values for source waters.



**Figure S4.** Canonical correlation analysis relating qualitative DOM properties (SUVA, the six FDOM components, and  $S_{285}$ ; red) to relative proportions of THMs and HAAs (TCM, BDCM, DBCM, DBAA, DCAA, TCAA, MBAA, BCAA; blue) showing the variables (A) and the hamlets by year (B).