



## Diffusive fluxes of persistent organic pollutants between Arctic atmosphere, surface waters and sediments



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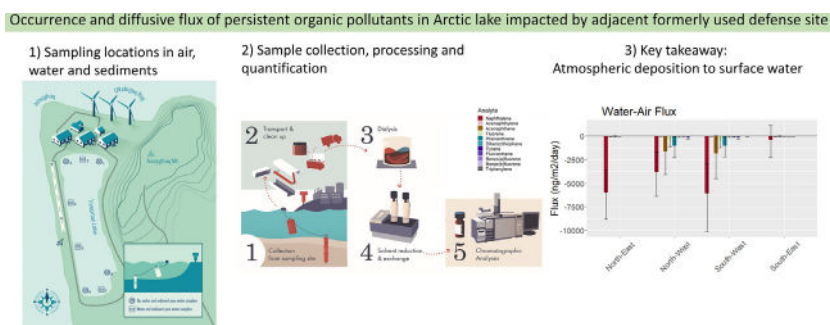
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### HIGHLIGHTS

- Atmospheric deposition was the primary route of chemical movement in Troutman Lake.
- PAH and PCB concentrations were low and comparable to other remote/rural locations.
- Tris(2-chloroethyl)phosphate was higher than previously reported for Arctic atmosphere.
- Demonstrates relevance of modern and legacy sources emitted both globally and locally

### GRAPHICAL ABSTRACT



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### ABSTRACT

Arctic communities are disproportionately exposed to pollutants from sources including global atmospheric transport and formerly used defense sites (FUDS). The effects of climate change and increasing development in the Arctic have the potential to exacerbate this problem. Yupik People of Sivuqaq, or St Lawrence Island, Alaska are one such community with documented exposures to pollutants from FUDS, and their traditional lipid-rich foods such as blubber and rendered oils of marine mammals. Troutman Lake, adjacent to the Yupik community of Gambell, Alaska, was used as a disposal site during the decommission of the adjacent FUDS, leading to community concern about exposure to military pollution and intrusion from historic local dump sites. In collaboration with a local community group, this study utilized passive sampling devices deployed in Troutman Lake. Air, water and sediment deployed samplers were analyzed for unsubstituted and alkylated polycyclic aromatic hydrocarbons (PAHs), brominated and organophosphate flame retardants and polychlorinated biphenyls (PCBs). PAH concentrations were low and comparable to other remote/rural locations. PAHs were generally in deposition from the overlying atmosphere into Troutman Lake. Of the flame retardants, brominated diphenyl ether-47 was detected in all surface water samplers while triphenyl phosphate was detected in all environmental compartments. Both were at concentrations equivalent or lower than other remote locations. Of particular interest, we measured higher atmospheric concentrations of tris(2-chloroethyl) phosphate (TCEP) (0.75–2.8 ng/m<sup>3</sup>) than previously reported in the literature for remote Arctic sites (<0.017–0.56 ng/m<sup>3</sup>). TCEP was found to be in deposition to Troutman Lake at magnitudes from 290 to 1300 ng/m<sup>2</sup>/day. No PCBs were detected in this study. Our findings demonstrate the relevance of both modern and legacy chemicals from local and global sources. These results help us to understand the fate of anthropogenic contaminants in dynamic Arctic systems providing valuable data for communities, policy makers and scientists.

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## 1. Introduction

Persistent organic pollutants (POPs) pose a unique hazard to Indigenous peoples living in the Arctic. Indigenous peoples in the far north are disproportionately exposed to contaminants as a result of global atmospheric transport, the abundance of current and former military facilities, and traditional subsistence diets rich in marine mammals which are known to contain high levels of many POPs due to biomagnification (Arctic Monitoring and Assessment Programme, 2012). Global climate change has the potential to exacerbate these issues. The effects of climate change are magnified at extreme latitudes and altitudes making the Arctic an important locus for understanding these impacts. Changes in precipitation, temperature driven changes in chemical partitioning, and melting sea ice, glaciers and permafrost all pose challenges to assessing the movement of chemicals in the Arctic (Macdonald et al., 2003). Other impacts of climate change such as altered species distribution and trophic transfer are poorly understood, yet have caused important changes in food web mediated exposure to POPs (Borgå and McKinney, 2022; Moe et al., 2013). A growing body of literature has demonstrated the release of POPs from melting sea ice, glaciers, and permafrost yet many knowledge gaps remain about the effect of global climate change on contaminant movement in the cryosphere (Hung et al., 2022; Ma et al., 2011; Ren et al., 2019).

Sivuqaq lies south-west of Nome in the northern Bering Sea approximately 60 km from the Siberian Chukchi Peninsula. The island is home to Sivuaq Yupik People who practice a traditional subsistence lifestyle, harvesting edible plants, berries, birds, fish and marine mammals (Miller et al., 2013). During the Cold War, two military installations were developed on Sivuaq due to its proximity to Russia. One installation was at the Native Village of Gambell and upon closure in the 1950's debris and chemical waste were buried by the military in disposal sites around the village and in the adjacent Troutman Lake. Despite the use of the lake by the military as a disposal site for hazardous waste, there has been little investigation into chemical contaminants in the lake conducted by the Army Corps of Engineers (USACE, 2005). A recent study conducted by Zheng et al. revealed the occurrence of polychlorinated biphenyls, brominated and organophosphate flame retardants in addition to per/polyfluorinated compounds in the tissues of resident fish in Troutman Lake (Zheng et al., 2020). However, the source of these chemicals and their abundance in the abiotic compartments of the lake is not well understood. In addition, several studies have demonstrated that some flame retardants may be subject to long range transport demonstrating the importance of research to better understand the interplay between point source and global transport of these POPs in Troutman Lake and across the Arctic (Breivik et al., 2006; Möller et al., 2012; Prats et al., 2022a; Sun et al., 2020).

One effective tool to address these knowledge gaps is passive sampling. Passive sampling devices (PSDs) have been previously established as a sampling technique well suited to measuring trace levels of organic pollutants in a variety of environmental matrices. PSDs are particularly well suited to sampling in remote environments where logistical constraints prevent the application of other sampling techniques (Yao et al., 2016; Lohmann and Muir, 2010; Prats et al., 2022b). PSDs accumulate chemicals from the freely dissolved or vapor phase and as a result can provide a highly sensitive measure that reflects the time weighted average concentration, capturing episodic temporal variations. Low-density polyethylene (LDPE) is a lipophilic polymer capable of sampling semi-volatile organic compounds with a wide range of physico-chemical properties facilitating simultaneous investigation of several compound classes (Vrana et al., 2005). Using passive sampler derived concentrations in models of diffusive flux allow us to determine the direction and magnitude of chemical movement between environmental compartments, improving our understanding of pollution sources and chemical transport (Ruge et al., 2015; Prats et al., 2023; Khairy and Lohmann, 2017). We demonstrate the utility of pairing passive sampling and diffusive flux models to determine the movement of chemicals in a remote Arctic environment. To our knowledge this represents the first use of passive samplers to describe diffusive flux of persistent organic pollutants between sediments, surface water and atmosphere in the Arctic.

Responsive to community concerns, this study describes the occurrence and movement of flame retardants, PCBs and PAHs between air, water and sediments none of which had previously been measured in Troutman Lake and are rarely monitored in remote Arctic communities. These compound classes were selected due to documented releases of fuels and PCB containing materials and community interest in flame retardant exposures. We use flame retardant and PAH fugacity to better understand the dynamic contributions of point sources and global transport in remote Arctic regions where climate change driven processes affect chemical movement and Indigenous Peoples of the North.

## 2. Materials and methods

### 2.1. Study area

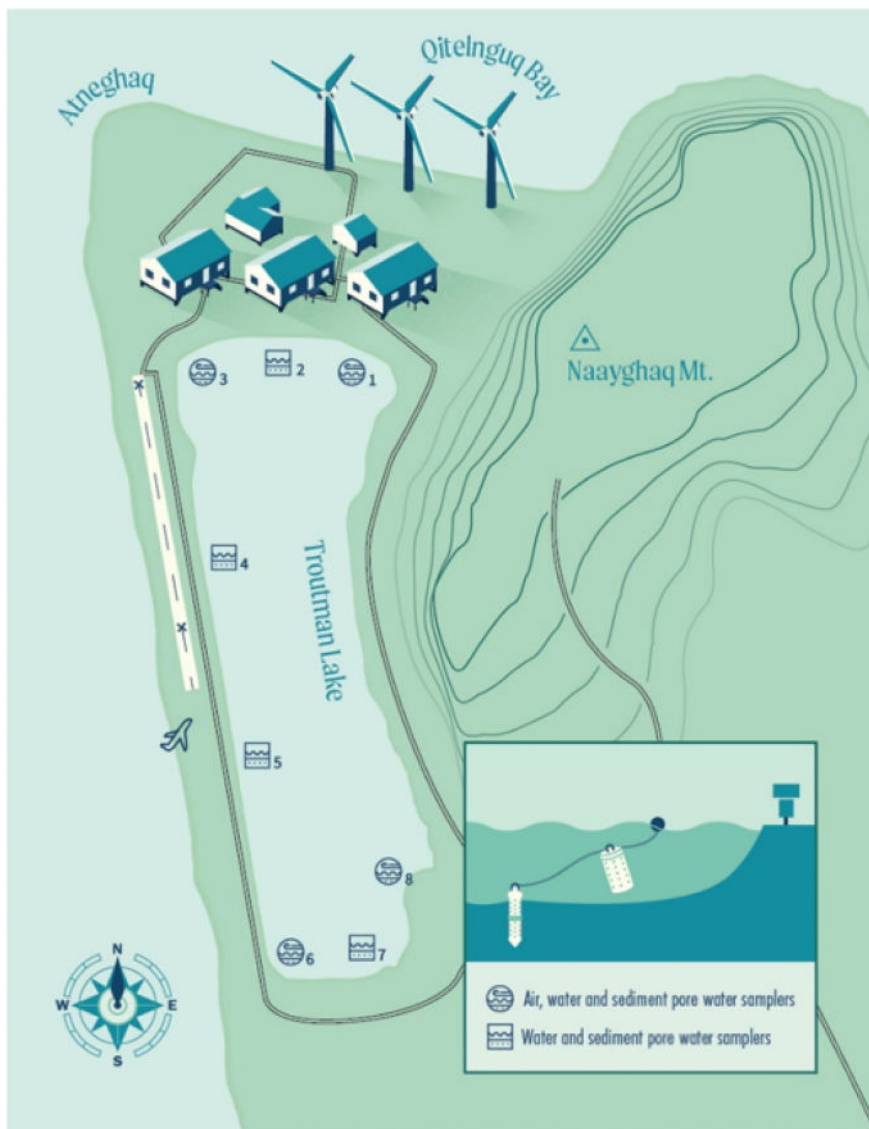
Troutman Lake lies directly south of the village of Gambell on the north-west corner of Sivuaq. Approximately three-square kilometers, the lake is bordered by an airstrip and rocky beach to the west, ephemeral wetlands to the south and Naayghaq Mountain to the east. During remediation 38 sites of concern were identified by the Army Corps of Engineers including military landfills, discarded chemical storage drums, munitions and electrical transformers (USACE, 2005). In particular, the north side of Troutman Lake was used as a disposal site by the US military. Investigations into reports of munitions in the lake revealed metal debris and 55-gal drums, the contents of which are unknown (USACE, 2005). In response to community concerns, in 2017 an Agency for Toxic Substances and Disease Registry (ATSDR) health consultation was released which determined the safety of recreation in Troutman Lake was uncertain due to insufficient characterization of chemical contamination. The extent of characterization of chemical contaminants in Troutman Lake is a single surface water sample collected by the US Army Corps of Engineers in 1985 and analyzed for water quality parameters described in the ATSDR health consultation for Gambell FUDS (Gambell\_FUDS\_HC\_092020-508.Pdf, n.d.).

### 2.2. Sampler deployment

PSDs were prepared as described in detail elsewhere (Anderson et al., 2008). PSDs were deployed in and around Troutman Lake in the air, water and sediment pore water (referred to hereafter as sediment) in July 2019. Air samplers were deployed within 5 m of the shoreline at 4 locations (North-East, North-West, South-West and South-East corners of the lake) and water and sediment samplers were deployed at 8 locations along the north, west and south sides of the lake (Fig. 1). Air samplers consisted of 5 low density polyethylene (LDPE) strips housed in a sheet metal box with open ends. Surface water samplers utilized 5 LDPE strips deployed in a perforated steel cage while samplers consisted of a single strip of LDPE housed in a perforated steel probe driven into the sediment (Minick and Anderson, 2017). Triplicate samplers were deployed in air and water at the north-west corner of the lake. Each water sampler was deployed approximately 1 m deep, suspended on a cable between a weight and a floating buoy. Sediment probes containing PSDs were collocated with each water sampler and placed in the sediment with the top of the probe flush with the sediment-water interface and the middle of the sampler approximately 16-17 cm deep. Samplers were deployed for 7 days, collected and returned to Oregon State University for extraction and analysis. Upon return to the laboratory, samples were stored at  $-20^{\circ}\text{C}$  prior to cleaning and extraction (Donald et al., 2016).

### 2.3. LDPE cleaning and extraction

PSDs were cleaned to remove biofouling and extracted as described elsewhere (Anderson et al., 2008; Huckins et al., 2006). Prior to extraction, samplers were spiked with surrogate standards to assess recovery across the extraction process. PSDs were extracted by two rounds of hexane dialysis. Solvent extracts were reduced to 1 mL using a TurboVap closed cell evaporator. Sample aliquots were spiked with internal standards for the respective analytical methods as applicable. All solvents used were Optima grade or better.



**Fig. 1.** Location of passive sampling devices deployed in Troutman Lake. Site names used in manuscript provided along with local place names. 1) North-East (Maqneghluqaq), 2) Center-North (Tapeghaq), 3) North-West (near Pitegseghaghviyet), 4) Airstrip-North (South of Tekeghaghpak), 5) Airstrip-South (Naayvam Ketlighnegha), 6) South-West (near Qamughtut), 7) Center-South (Naayvam Kenlenga), 8) South-East (near Siighmiik). See SI Table S1 for sampling location GPS coordinates.

#### 2.4. Instrumental analysis

Samples were analyzed for 41 unsubstituted and 22 alkylated polycyclic aromatic hydrocarbon (PAH) targets by gas chromatography-triple quadrupole mass spectrometry (GC-TQMS). The analysis was conducted using an Agilent 7890A GC and an Agilent 7000C tandem mass spectrometer using the method described in Anderson et al. 2015 (Anderson et al., 2015). Analysis for 52 polychlorinated biphenyls (PCBs) utilized an Agilent 6890 M GC with electron capture detector. An Agilent 7890A GC and an Agilent 5975C mass spectrometer were used to measure 44 organophosphate and brominated diphenyl ether flame retardants (OP and BDE, respectively). Instrument parameters and details on internal standards, surrogates, targets and detection limits for each analysis can be found in supplemental information (SI) section 3. PAH surrogate recoveries ranged from 38 % to 78 % with an average recovery of 57 %. Surrogate recoveries for PCB analysis ranged from 58 % to 77 % with an average recovery of 67 %. PAH concentrations were surrogate corrected to account for loss during the extraction process.

#### 2.5. Quality control

A variety of quality control (QC) samples were used to ensure sample integrity during PSD construction, deployment, retrieval, transportation and laboratory processing. No targets were detected in any instrument blanks and check standards confirmed instrument calibration in all cases. No PCBs or flame retardants were detected in any QC samples. Low levels of some PAHs were detected in QC samples. The average of laboratory QC sample concentrations were used for background subtraction. Targets detected in QC samples and background subtracted concentrations can be found in Table S7. Samplers deployed in triplicate had an average RSD of 37 % and 27 % for PAHs and flame retardants, respectively, for compounds above the limit of quantitation. Duplicate aliquots were analyzed for PAHs and flame retardants. The average relative percent difference between duplicate aliquots was 13 % for PAHs. No flame retardant targets were detected in either of the duplicate aliquots. Matrix spike samples were analyzed for PAHs and flame retardants and the average recovery was 63 % and 88 %, respectively.

2.6. Calculations

Time weighted average concentrations in air, water and sediment were calculated using the empirical uptake model (Huckins et al., 2006). Depuration of performance reference compounds (PRCs) were used to calculate in-situ sampling rates. Detailed equations are provided in SI section 2.

Diffusive flux was calculated using models described in detail elsewhere (Minick and Anderson, 2017; Tidwell et al., 2016; Bamford et al., 1999). Briefly these models use derivations of Fick's first law in which the concentration gradient between two compartments is multiplied by a mass transfer coefficient to derive the diffusive flux of targets across the air-water and sediment-water interfaces (See SI section 2.2 and 2.3 for equations). Uncertainties in calculated flux were described using two complementary approaches, a Monte Carlo simulation approach and the propagation of error approach described in detail elsewhere (Minick and Anderson, 2017; Liu et al., 2016). Briefly both approaches use observed variability in replicate samplers and assumed uncertainty in mass transfer coefficients and Henry's Law constants to describe the uncertainty in estimates of diffusive flux (Additional details available in SI section 2.4). Standard deviation equivalents were derived from the relevant quantiles of the Monte Carlo simulated distribution of flux values.

2.7. Data sharing

With our partners, the leadership of Sivuqaq and Alaska Community Action on Toxics (ACAT), we implemented a policy around data sharing. The policy stipulates that data must be reviewed by the Sivuqaq Working Group, and the Native Village of Gambell Tribal Council prior to dissemination. Additionally, community partners are integrally involved in presentations and publications. A six-page summary report was developed, highlighting the results of air, water and sediment sampling across three chemical categories: polycyclic aromatic hydrocarbons, polychlorinated

biphenyls, and flame retardants. The report was developed following guidelines from the Clear Communication Index (The CDC, n.d.) and written at an 8th grade reading level or lower. A cover letter described the background and rationale of the study, including background on the work done by ACAT. Community questions were listed in parallel to the results of the data to provide a high-level overview of how the data were interpreted. A graphical map identified the general locations of the samplers. Subsequent pages described the levels of chemicals in and around Troutman Lake, and comparisons to other areas of the US were provided as context. Information on the passive samplers was provided in an infographic. The report was internally developed and reviewed, and then reviewed by ACAT, the Sivuqaq Working Group, and the Native Village of Gambell Tribal Council. Following final changes and approval, ACAT provided the report to the Tribe and other stakeholders. (See SI for report back materials).

3. Results and discussion

3.1. PAHs

3.1.1. Air, water and sediment concentrations

Unsubstituted PAHs, referred to hereafter as PAHs, were detected in all air samplers with sum PAH concentrations ranging from 18 to 40 ng/m<sup>3</sup>. Higher concentrations were observed on the west side of the lake compared to the east side of the lake (Fig. 2). The distribution of individual PAHs was similar between sites. Vapor phase PAHs were predominantly two and three ring compounds (92 % and 8 %, respectively). Four ring PAHs represented <1 % of the total in all cases. No PAHs were detected in the air with greater than four rings. In the surface water the PAH composition and concentrations were relatively homogenous with sum PAH concentrations ranging from 1.4 to 3.2 ng/L. Similar to vapor phase results, naphthalene was the most abundant compound in the surface water composing 92 % and 87 % of the measured PAH concentrations in the air and water,

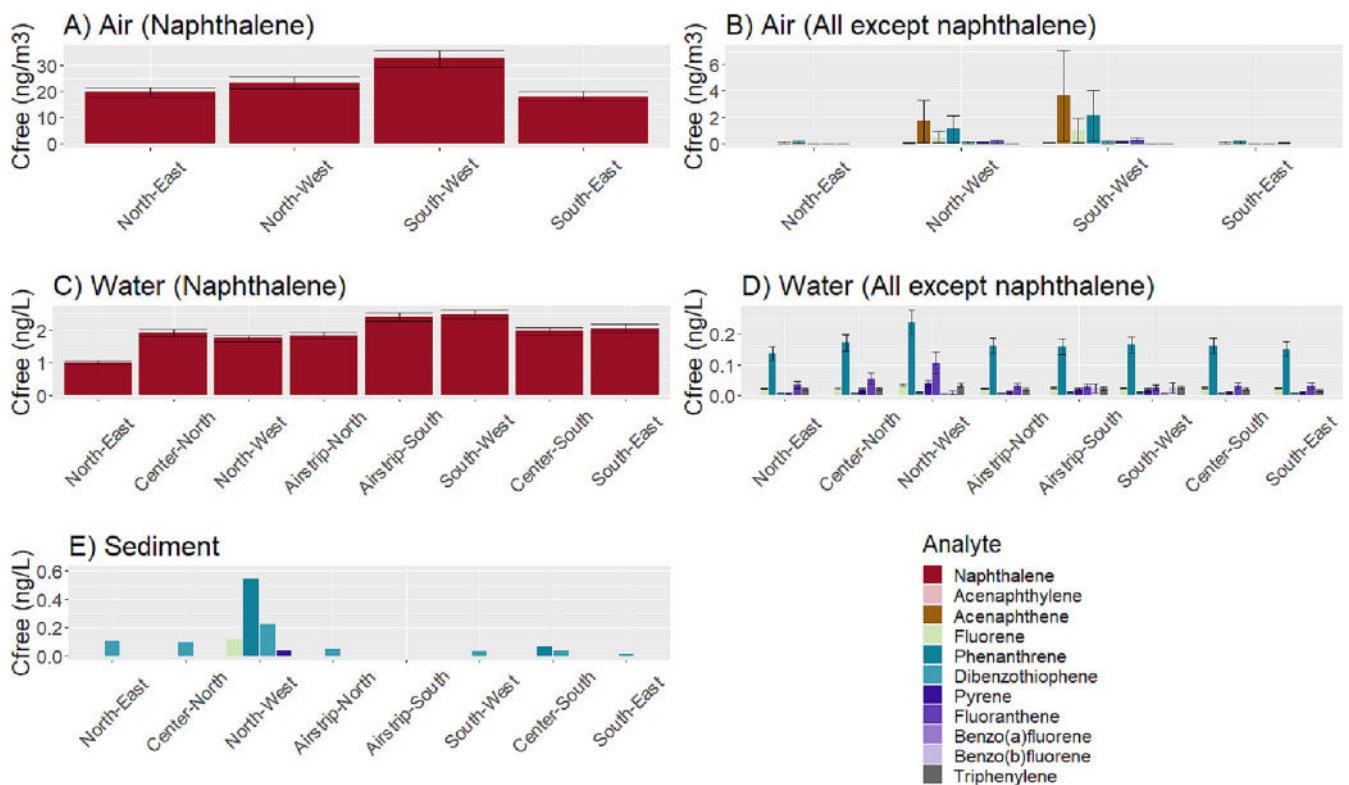


Fig. 2. Concentrations of unsubstituted PAHs in air (A & B), water (C & D), and sediment (E). Concentrations were generally low with naphthalene constituting the majority of measured compounds in air and water while sediment detections were primarily three ringed PAHs. Error bars represent standard deviation of triplicate field deployed samplers.

respectively. Sediment concentrations were generally lower than surface water concentrations with sum PAH concentrations ranging from all targets below detection limits to 0.93 ng/L. Greater heterogeneity was observed in the sediment samplers relative to surface water samplers with markedly higher concentrations at North-West sampling location (0.93 ng/L compared to 0.10 ng/L at the next highest site). Sediment samplers had fewer detections and lower concentrations than surface water samplers. In contrast to surface water and air measures, sediment contained predominantly three ring PAHs, especially phenanthrene and sulfur containing heterocycle dibenzothiophene. Pyrene was the only four ring PAH detected in sediment samplers, constituting <5 % of the total. Naphthalene was not detected in any sediment samples.

Alkylated PAHs displayed similar patterns, with alkylated naphthalenes composing the majority of measured alkyls in air and water (Fig. 3). Sum of alkyl PAHs ranged from 2 to 15 ng/m<sup>3</sup> in the air and 0.15 to 0.42 ng/L in surface water. In the sediment only alkylated naphthalenes and phenanthrene were detected. At three of the eight sites all targets were below detection limits. The highest concentrations were measured in the sediment at Center-North where the sum of alkyls was 0.42 ng/L. Unsubstituted and alkylated PAHs were correlated with each other in the air and water, with the highest concentrations of both unsubstituted and alkylated PAHs measured at the South-West corner of the lake.

The concentrations observed in this study are similar to results from other rural locations and are in concordance with modern anthropogenic inputs (Fig. 4) (Minick and Anderson, 2017; Tidwell et al., 2016; Ghetu et al., 2022; Paulik et al., 2016; Minick et al., 2019; Villeneuve et al., 2005; Allan et al., 2012). Combustion is the largest emission source of vapor phase naphthalene (Jia and Batterman, 2010). Potential local sources include diesel combustion in the community power plant, burning stove oil for heating residential homes, gasoline powered vehicles and cigarette smoke. The range of vapor phase sum unsubstituted and alkylated PAH concentrations measured here are slightly lower than the range of values reported for rural communities around the Pacific Northwest during the

summer in 2019–2022 (37–293 ng/m<sup>3</sup>) (Ghetu et al., 2022). Similarly, surface water concentrations reported in this study are comparable to the concentrations measured in the Willamette River, Oregon, upriver of major industrial sources in Corvallis and Newberg in 2003 (3.22 ng/L +/- 1.22 (SE, n = 2) and 1.72 ng/L +/- 0.54, respectively) (Villeneuve et al., 2005). The sediment concentrations measured here are lower than the values reported in Minick et al. 2019 which measured sediment pore water concentrations at tribal clamming beaches in Washington State (1.2–15 ng/L) (Minick et al., 2019) and lower than measures in the Willamette River (Minick and Anderson, 2017).

### 3.1.2. Diffusive flux

Across the air-water interface PAHs were found to be predominantly in deposition throughout the lake. The greatest magnitude of depositional flux was on the west side of the lake, corresponding to the higher measured vapor phase concentrations at the North-West and South-West sample sites. The sum of depositional fluxes ranged from 16,000 ng/m<sup>2</sup>/day at the South-West sample site to 200 ng/m<sup>2</sup>/day at the South-East location (Figs. 5, 6). The number of compounds in deposition, when accounting for uncertainty, ranged from 1 to 15 with the most compounds in deposition at the South-West sample site. The compounds with the largest depositional magnitude were naphthalene and alkylated derivatives of naphthalene. The only instance of volatilization was 1,2-dimethyl naphthalene at South-West which had a magnitude of 93 ng/m<sup>2</sup>/day. The magnitude of atmospheric inputs of naphthalene and its alkylated derivatives suggest that the water concentrations we observed are a result of and share a common source with the measured vapor phase compounds rather than migration from the sediments.

The majority of diffusive flux across the sediment-water interface was naphthalene in deposition from the overlying surface water (Fig. 4.B). The next largest contributor to deposition was phenanthrene. Diffusive release from sediment into surface water was of lower magnitude and included dibenzothiophene and various alkylated naphthalenes. The low

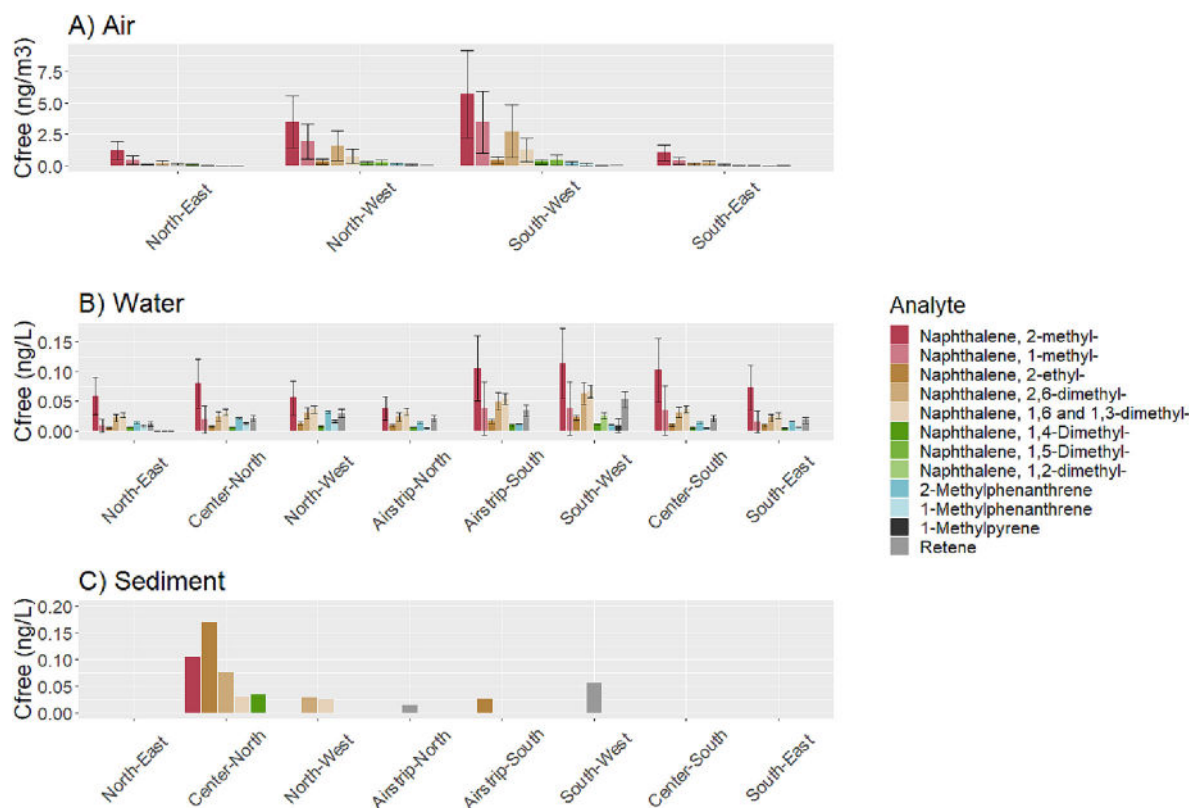


Fig. 3. Concentrations of alkylated PAHs in air (A), water (B), and sediment (C). Concentrations were generally low with alkylated naphthalenes constituting the majority of measured compounds. Error bars represent standard deviation of triplicate field deployed samplers.

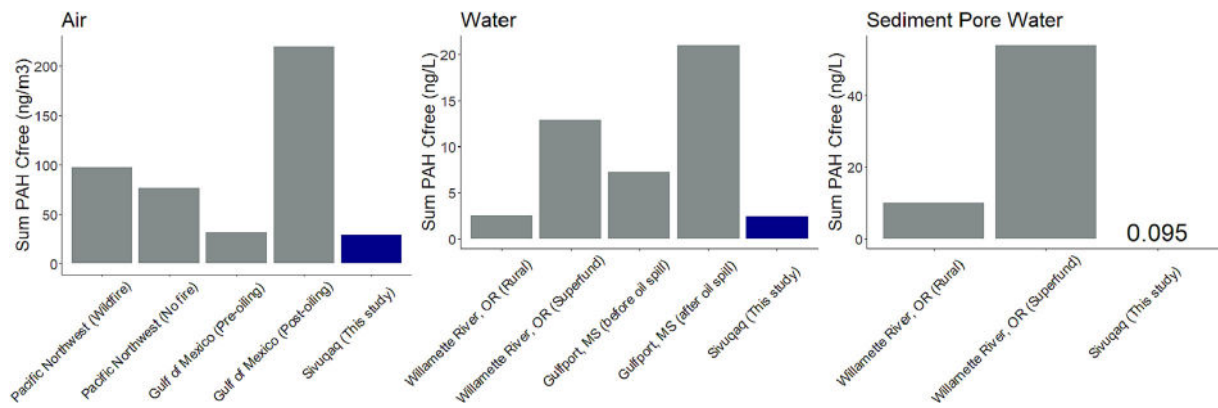


Fig. 4. Sum PAH concentrations measured in this study were equivalent or lower than values measured elsewhere at rural or unimpacted sites in North America (Vrana et al., 2005; Gambell\_FUDS\_HC\_092020-508.Pdf, n.d.; Huckins et al., 2006; Anderson et al., 2015; Tidwell et al., 2016; Bamford et al., 1999; Liu et al., 2016).

magnitudes of diffusive flux across the sediment-water interface reflect the small differences in concentration between the compartments. This is likely a result of mixing between water and sediment pore water due to the high permeability of the gravel lakebed (Vervier et al., 1992).

### 3.2. Flame retardants

#### 3.2.1. Air, water, sediment concentrations

Two flame retardants were detected in all air samplers, tris(2-chloroethyl) phosphate (TCEP) and triphenyl phosphate (TPP), with a range of concentrations of 0.75–2.8 ng/m<sup>3</sup> and 0.001–0.004 ng/m<sup>3</sup>, respectively. Higher concentrations were observed at the north end of the lake compared to the south (Fig. 7). Two flame retardants were found in all

water samplers, TPP and BDE-47. TPP was the more abundant of the two compounds with concentrations from 0.017 to 0.07 ng/L. BDE-47 was present in all water samplers between 0.002 and 0.016 ng/L. Only TPP was detected in the sediment and only at two locations, North-East and Airstrip-South at 0.038 and 0.065 ng/L, respectively.

These results are in concordance with a previous study investigating flame retardant concentrations in fish in Troutman Lake. The two flame retardants detected in the surface water in this study, triphenyl phosphate and BDE-47, were detected in fish tissue in Troutman Lake by Zheng et al. 2020 (Zheng et al., 2020). While other flame retardants were detected by Zheng et al., BDE-47 was found to be the most abundant BDE congener present in fish tissue. The previous detection of additional compounds in fish not detected in surface waters in the current study is likely a result of

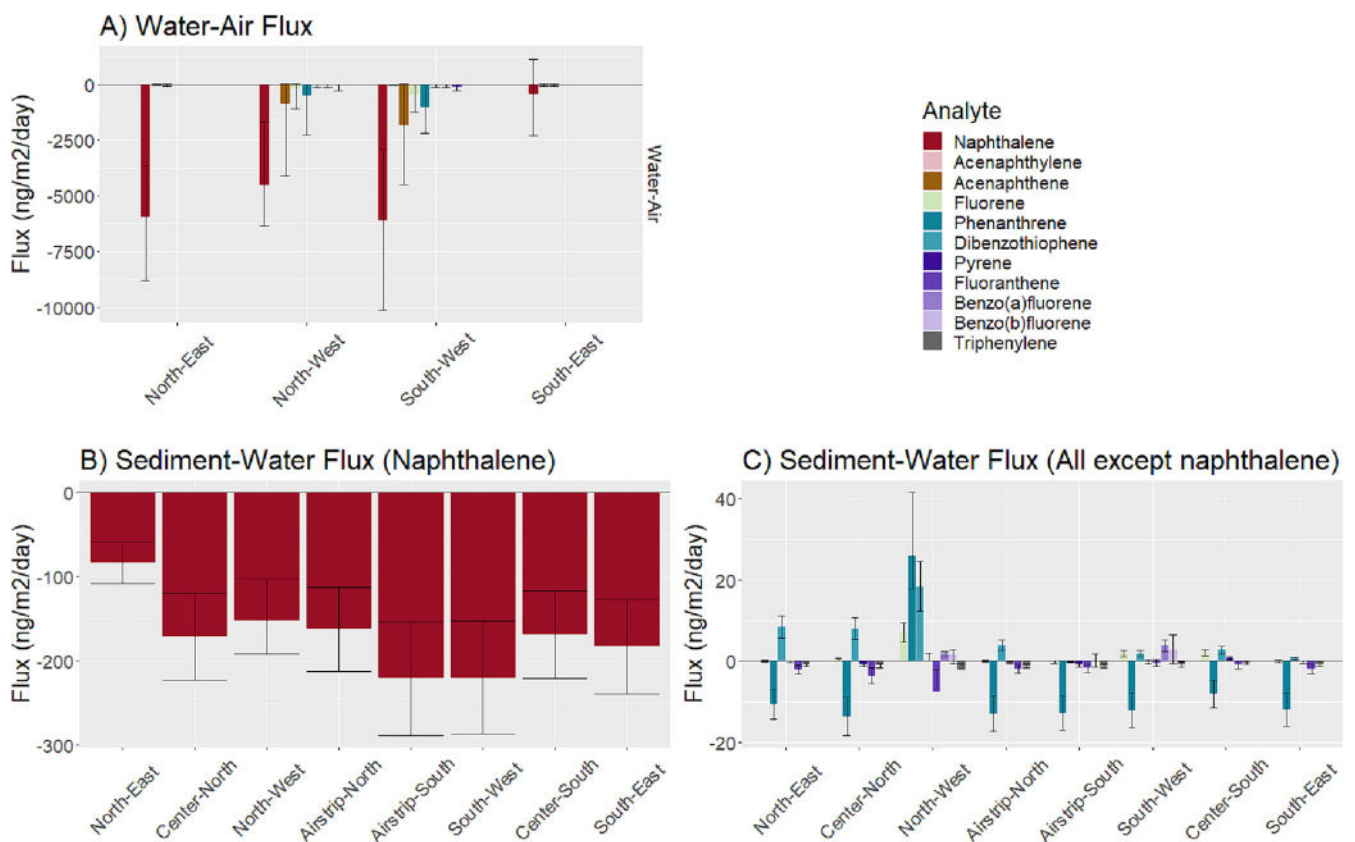
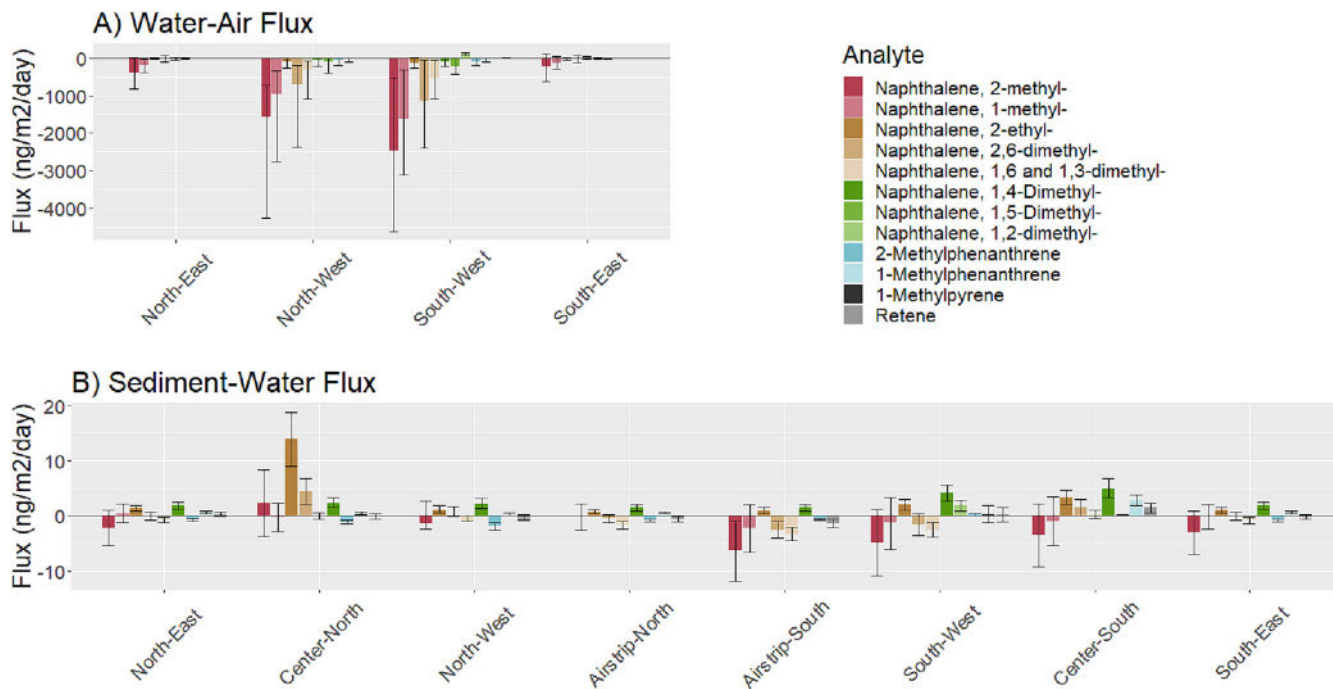


Fig. 5. Fluxes of unsubstituted PAHs across the air-water (A) and sediment-water (B & C) interfaces were predominantly in deposition with naphthalene comprising the majority of chemical movement. Error bars represent prediction intervals from Monte Carlo simulations corresponding to plus or minus one standard deviation (68 %) accounting for replicate variability and uncertainty in model parameters as described in supplemental information.

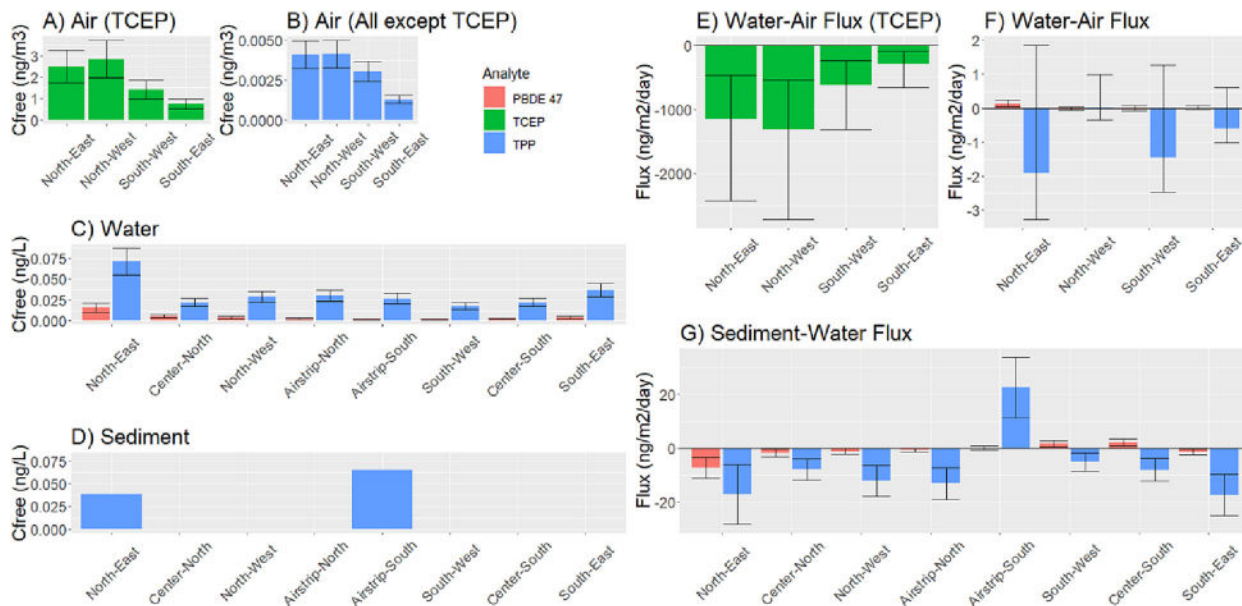


**Fig. 6.** Fluxes of alkylated PAHs across the air-water interface (A) were predominantly in deposition with alkylated naphthalenes comprising the majority of chemical movement. Across the sediment-water interface (B) there was a mix of deposition and release, primarily of alkylated naphthalenes. Error bars represent prediction intervals from Monte Carlo simulations corresponding to plus or minus one standard deviation (68 %) accounting for replicate variability and uncertainty in model parameters as described in supplemental information.

their bioconcentration into fish and the limited sampling duration of seven days. Compared to other studies in the literature, the surface water concentrations of BDE-47 and TPP measured in this study are comparable to those reported by McDonough et al. 2018 in marine Arctic surface waters (Below detection limit (BDL)-0.015 ng/L and 0.001–0.04 ng/L for BDE-47 and TPP, respectively) (McDonough et al., 2018).

One challenge to contextualizing atmospheric concentrations of flame retardants is methodological differences between studies. Several previous studies have reported data on particulate phase measurements as many

OPFRs are expected to be primarily associated with atmospheric particulates (Möller et al., 2012; Sühring et al., 2016a; Salamova et al., 2014; Ren et al., 2016). However, OPFRs have a wide range of physico-chemical properties and many are predominately in the vapor phase, including TCEP (Sühring et al., 2016b; Wang et al., 2020). Relative to other studies measuring vapor phase concentrations of TPP in the Arctic (0.01–0.25 ng/m<sup>3</sup>), the concentrations measured here are more than an order of magnitude lower (Fig. 8) (Rauert et al., 2018). Concentrations of TCEP are higher than vapor phase concentrations measured outdoors in



**Fig. 7.** A through D depict freely dissolved concentrations of brominated and organophosphate flame retardants in the air (A and B), water (C) and sediment (D). Error bars represent standard deviation of triplicate field deployed samplers. The remaining panels represent diffusive fluxes of brominated and organophosphate flame retardants across the air-water (E and F) and sediment-water (G) interfaces. Error bars represent prediction intervals from Monte Carlo simulations corresponding to plus or minus one standard deviation (68 %) accounting for replicate variability and uncertainty in model parameters as described in supplemental information.

urban environments (0.032–0.60 ng/m<sup>3</sup>) (Peverly et al., 2015; Saini et al., 2019). Studies reporting particulate phase or combined vapor and particulate phase concentrations have reported mean values as high as 10.5 and 14.6 ng/m<sup>3</sup> in Shanghai, China and Shimizu, Japan, respectively (Ren et al., 2016; Ohura et al., 2006). However, the concentrations measured in this study are higher than any values reported in the literature for similar remote Arctic sites (BDL–0.56 ng/m<sup>3</sup>) (Rauert et al., 2018). During the summer of 2014 passive samplers were deployed near the Native Village of Savoonga, located 63 Km from Troutman Lake, as part of the Global Atmospheric Passive Sampling Network. That study reported TCEP below detection limit (<0.017 ng/m<sup>3</sup>), one to two orders of magnitude lower than the concentrations reported in the current study (Rauert et al., 2018). While long range transport of chlorinated organophosphates is described in the literature, the magnitude of the values measured in this study compared to other comparable Arctic sites suggest local point sources may contribute to vapor phase TCEP concentrations around Troutman Lake (Möller et al., 2012; McDonough et al., 2018; Rauert et al., 2018; Li et al., 2017). While commercially developed in the mid 1960's the majority of use of organophosphate flame retardants like TCEP has been in the last twenty years as a replacement for brominated flame retardants, which were phased out in the early 2000s (Chupeau et al., 2020). The occurrence of these organophosphate flame retardants is unlikely to be associated with the FUDS as the defense site precedes the use of these chemicals. TCEP is widely used as a flame retardant in polyurethane foam, upholstery and building materials and has been found in high concentrations in the air around landfills in other studies (Kerric et al., 2021; Final Scope of the Risk Evaluation for Tris(2-Chloroethyl) Phosphate (TCEP), n.d.). Local waste disposal practices and a nearby open-air landfill may represent an important source of atmospheric TCEP concentrations in this area. While the landfill is 900 and 1400 m from the North-West and North-East sampling locations, respectively, the concentrations reported in this study are very similar to concentrations reported by Kerric et al. 2021 which described a mean combined vapor and particulate phase concentration of 2.4 ± 0.4 (SE) ng/m<sup>3</sup> around a municipal landfill near Montreal, Canada (Kerric et al., 2021). Waste management practices in the Arctic are the product of unique logistic and socioeconomic constraints (Burns et al., 2021). In some cases, landfill conditions have resulted in hazardous emissions to residents of other Arctic communities, however there are few data on emissions from waste management practices in the Arctic (Weichenthal et al., 2015).

### 3.2.2. Diffusive flux

The largest contributor to diffusive flux was the deposition of TCEP from the atmosphere into the surface water (Fig. 7E). Deposition of TCEP was higher at the north end of the lake than the south end of the lake,

corresponding to the higher air concentrations measured on the north side of the lake (Fig. 7A). The most deposition occurred at the North-West site with a magnitude of 1300 ng/m<sup>2</sup>/day (Fig. 7E). The least diffusive flux of TCEP across the air-water interface occurred at the South-East sample site at a rate of 290 ng/m<sup>2</sup>/day. BDE-47 and TPP showed little movement between air and water with all locations showing <2 ng/m<sup>2</sup>/day (Fig. 7F).

The direction of diffusive flux at the water-sediment interface was primarily deposition from the surface water (Fig. 7G). TPP had a larger magnitude in all cases and ranged from 17 ng/m<sup>2</sup>/day to 5.2 ng/m<sup>2</sup>/day for deposition into sediment. TPP was in release from the sediment at only one site, Airstrip-South with a magnitude of 22 ng/m<sup>2</sup>/day. At most sites BDE-47 was in deposition into the sediment with magnitudes ranging from 0 to 7.3 ng/m<sup>2</sup>/day. Overall, the magnitude of diffusive flux across the sediment-water interface was very low as a result of the low concentrations and small gradient measured in the surface water and sediment (Fig. 7C and D).

### 3.3. PCBs

No PCBs were detected in any samples. See table S4 for a full list of target analytes and detection limits. PCBs were previously detected in fish in Troutman Lake by Zheng et al. which found hexachlorinated biphenyls to be the most abundant congeners. As previously mentioned, compounds detected in fish by Zheng et al., not detected in the present study are likely the result of bioconcentration in fish, and therefore not captured in the relatively short, seven day passive sampling duration.

## 4. Conclusions

To our knowledge this study represents the first use of passive sampling devices combined with diffusive flux models to describe the movement of chemicals between air, surface water and sediment in remote arctic environments. While we did not observe evidence of chemicals in release from military landfills with melting permafrost or re-volatilization of historic sinks, this work demonstrates the utility of passive samplers to address the challenges to assessing chemical fate in the era of climate change. Air-water partitioning of semi-volatile organic chemicals is highly temperature sensitive and as temperatures increases, the direction and magnitude of diffusive flux across the air-water interface may shift dramatically. Studies like this one can provide site specific information about the movement of chemicals to improve our understanding of chemical fate in the Arctic as climate change continues to affect these systems. The implication of local, modern sources is in line with a growing body of literature recognizing

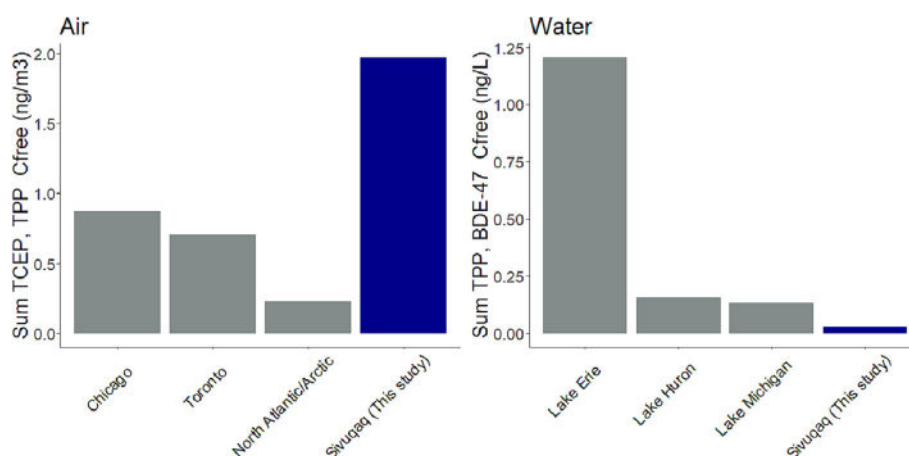


Fig. 8. Median sum concentrations of flame retardant detected in this study were higher than published vapor phase values in the atmosphere but lower than published surface water concentrations. Data presented here are limited to vapor phase measures, see Wong et al. 2018 and Li et al. 2017 for comparisons with particulate only and combined vapor and particulate concentrations (Li et al., 2017; Saini et al., 2019; Salamova et al., 2014; Wong et al., 2018). No comparison values available in the literature for sediment pore water.



the importance of evaluating both long range transport and local emissions to understand anthropogenic pollution in the Arctic (Schmale et al., 2018; Choi et al., 2008; Law et al., 2017). The data in this study can inform the Yupik People of Sivuqaq and other local, regional and global stakeholders in their efforts to protect human and ecosystem health in the Arctic.

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## CRediT authorship contribution statement

**Ian L. Moran:** Conceptualization, Formal analysis, Investigation, Writing – original draft, Visualization. **Lane Tidwell:** Conceptualization, Methodology, Resources, Investigation, Writing – review & editing. **Michael Barton:** Software, Data curation. **Molly Kile:** Conceptualization, Funding acquisition. **Pamela Miller:** Conceptualization, Project administration. **Diana Rohlman:** Visualization, Writing – review & editing. **Samarly Seguinot-Medina:** Conceptualization, Writing – review & editing. **Bobby Ungwiluk:** Conceptualization, Writing – review & editing, Investigation. **Vi Waghiyi:** Conceptualization, Writing – review & editing. **Kim Anderson:** Conceptualization, Methodology, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition.

## Data availability

Data will be made available on request.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2023.164566>.

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