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Evaluating mercury concentrations in edible plant and fungi species in the Canadian Arctic environment

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Abstract

Levels of environmental mercury (Hg) within the Canadian Arctic are a current area of concern. Although efforts have been made to reduce Hg released into the environment, levels remain elevated in flora and fauna. This study examined the concentrations of Hg in soil and naturally occurring edible plant and fungi species, identified by local Inuit residents, from eight locations in Iqaluit, Nunavut, and the surrounding area during the summers of 2018 and 2019. Total Hg concentrations were obtained in 24 soil samples, 112 flora samples from 23 plant and five lichen species, and 157 fungal samples from eight species. Median Hg concentrations in plant species ranged from 0.005 $\mu\text{g g}^{-1}$ Hg dry weight (dw) in *Saxifraga cernua* to 0.19 $\mu\text{g g}^{-1}$ Hg dw in *Oxytropis maydelliana*. Median concentrations in edible fungi species ranged from 0.084 $\mu\text{g g}^{-1}$ Hg dw in the *Cortinarius croceus* (non-puffball species) to 1.6 $\mu\text{g g}^{-1}$ Hg dw in *Lycoperdon perlatum* (a puffball mushroom). Additionally, median Hg concentration in puffball species (1.4 $\mu\text{g g}^{-1}$) were higher than non-puffball species (0.12 $\mu\text{g g}^{-1}$). Three puffball species were assessed for methylmercury (MeHg), with mean concentrations ranging from 0.013 to 0.085 $\mu\text{g g}^{-1}$ MeHg dw. Limited research has been conducted on Hg uptake in naturally occurring edible plant and fungi species of the Canadian Arctic. This study contributes important information on Hg accumulation and processes in edible plant and fungi Arctic species, is the first to focus on plants used by the local Indigenous community, and demonstrates a need for further studies to assess Hg in Arctic environments.

1 | INTRODUCTION

Mercury (Hg) is a naturally occurring element in the Earth's crust and can be harmful to humans and wildlife (Zhang & Wong, 2007). Mercury exists in several chemical forms, including elemental [Hg(0)], inorganic [Hg(II)], and organic forms such as MeHg and dimethylmercury (Me₂Hg), and can cycle through these forms as a result of biogeochemical processes in the environment (AANDC, 2012; USEPA, 2018).

Abbreviations: dw, dry weight; fw, fresh weight; GEM, gaseous elemental mercury.

Mercury from natural sources has remained relatively undisturbed for decades, but global cycling from anthropogenic sources, including fossil fuel emissions, waste incineration, chlor-alkali facilities, and mining and smelting processes, have led to a global contamination issue. It is estimated that atmospheric levels of Hg have increased 10-fold since pre-industrial times, with fossil fuel combustion and heat production contributing ~46% (1,920 t) of total Hg production worldwide annually (AANDC, 2012; AMAP, 2011). Mercury from sources worldwide is transported northward by atmospheric and upper oceanic currents and deposited in

the terrestrial and aquatic ecosystems of the Canadian Arctic (Brown et al., 2018). Local sources of Hg in the Canadian Arctic (e.g., weathering of rock, waste incineration, and fossil fuel combustion) contribute marginally compared with international anthropogenic sources (Durnford et al., 2010; Shoty, 2017). In 2005, Asia was the major contributor (65%) of all gaseous elemental Hg (GEM) [Hg(0)] in the Canadian Arctic, with Canada being responsible for <1% (AMAP, 2011). Although global Hg emissions have dropped 20% and GEM from anthropogenic sources have dropped by 30% since 1990, transport and deposit of global Hg to the Canadian Arctic continues (Zhang et al., 2016). Due to the concern of increasing levels of Hg, the Minamata Convention on Hg, an international treaty, has been designed to protect the environment and human health from anthropogenic sources of Hg and its negative effects and to tackle the increasing levels (UNEP, 2021).

Arctic conditions during the summer months (low temperatures and abundant sunlight) promote Hg deposition (mainly as GEM) onto Arctic environments (AANDC, 2012; Domergue et al., 2003; Poissant et al., 2008; Shoty, 2017). The concentration of Hg in the soils of the Arctic tundra in previous studies ranges from 0.046 to 0.111 $\mu\text{g g}^{-1}$, with median values up to 0.114 $\mu\text{g g}^{-1}$ (Gamberg et al., 2015; Halbach et al., 2017; Leitch, 2006; Loseto et al., 2004; Obrist et al., 2017; Olson et al., 2018), and the approximate annual net gain of 117 mg of Hg at the surfaces (land, ice, water) of the Arctic is evidence of the Arctic being a global sink of Hg (Dastoor et al., 2015; Obrist et al., 2017).

Mercury, as Hg(0), Hg(II), or MeHg, may enter a plant by being absorbed from the soil through the root system and translocated from the roots to the shoots and aboveground plant components. Mercury may also enter plant shoot components from the atmosphere via foliar absorption (e.g., diffusion from particulate, aqueous or gaseous forms across the cuticle/epidermis), with vapor/gaseous forms able to enter specifically through the stoma (AMAP, 1998; Azevedo & Rodriguez, 2012; Dombaiová, 2005; Kumar et al., 1995). Previous literature has demonstrated Hg accumulation via translocation in terrestrial plants, but most of the Hg absorbed from the soil remained in the root systems of the plants as Hg(0) and Hg(II) (Niu et al., 2011; Tangahu et al., 2011; Tomiyasu et al., 2005). Notably, Obrist et al. (2017) estimate that 90% of all Hg within Arctic plants is originally derived from atmospheric sources as GEM and demonstrate that levels in the terrestrial environment are enhanced during summer months from vegetative uptake. Through decomposition of plant material, Hg within vegetation perpetually accumulates within Arctic soils mainly in the form of Hg(II) (Gamberg et al., 2015).

Various studies have quantified Hg levels in plant and fungal species around the world (Azevedo & Rodriguez, 2012; Falandysz et al., 2012; Niu et al., 2011). In one study, rice (*Oryza sativa* L.) was grown in Hg-contaminated soils and

Core Ideas

- Mercury in the Canadian Arctic is at elevated concentrations in soils, plants, and fungi.
- Mercury levels in soils and 23 plant, five lichen, and eight fungal species were assessed.
- Levels of mercury in soils across locations were not statistically different.
- Concentrations ranged from 0.005 $\mu\text{g g}^{-1}$ Hg dw in *S. cernua* to 1.6 $\mu\text{g g}^{-1}$ Hg dw in *L. perlatum*.
- Fungi showed higher levels of mercury compared with plants, in particular puffballs.

was found to have concentrations of 8.0 $\mu\text{g g}^{-1}$ Hg wet weight (Eisler, 2006). Additionally, Hg levels in northeastern Poland and Spain mushroom species, including *Clitocybe rivulosa*, *Boletus edulis*, *Boletus pinophilus*, *Boletus aereus*, and *Boletus edulis*, have been found to range from 3.0 to 6.9 $\mu\text{g g}^{-1}$ Hg dry weight (dw) (Falandysz et al., 2003; Melgar et al., 2009; Nanda & Mishra, 1997). Mercury has been shown to enter fungi via ionic channels and to bind to sulfur and nitrogen ligands once inside. This binding is competitive with additional metals such as cadmium, zinc, and iron. According to Kojo and Lodenius (1989), Hg bioconcentration in fungi correlates with the amount of sulfhydryl-, methionine-, and disulfide-containing proteins because Hg binds to proteins and mimics certain compounds like methionine. Further, the vast mycorrhizal systems of certain fungi may bioconcentrate Hg from symbiotic plants (Fischer et al., 1995). The vast spread of the mycorrhizae networks and the mycelial longevity allows for wider distribution, with potential to accumulate large amounts of Hg over a longer period (ECCC, 2016).

In Canadian Arctic vegetation, lichens have been found to have high levels of total Hg compared with flowering plant species such as willows and sedges (AANDC, 2012; Choy et al., 2010; Gamberg 2009). Mercury accumulation in plant shoots of the Arctic is hypothesized to be a result of mainly atmospheric absorption rather than plant translocation. Several field and greenhouse studies have proven limited uptake of Hg from soils into the shoots because the roots act as a major Hg absorber rather than a transporter (Grigal, 2002; Shoty, 2017).

Arctic flora is of great importance to the Inuit because plant and fungal species are used by communities for natural consumption, materials for everyday use, fabrics, and medicinal remedies (Mallory & Aiken, 2012). Some plant and fungal species that are commonly used include the puffball (*Calvatia cretacea*), used for anti-septic and wound coagulant purposes; mountain sorrel [*Oxyria digyna* (L.) Hill], used to increase energy, to quench thirst, and to treat

arthritis; and the Arctic willow (*Salix arctica* Pall.), used for its anti-infection properties and to treat upset stomachs. Berry plant species, including crowberry (*Empetrum nigrum* L.), blueberry (*Vaccinium uliginosum* L.), cranberry (*Vaccinium vitis-idaea* L.), and cloudberry (*Rubus chamaemorus* L.), are vital traditional food items that provide high nutritional value for Inuit across the Arctic (Boulanger-Lapointe et al., 2019). Berries are harvested in significant quantities in August and September and are eaten raw immediately or are frozen for later use. Berries are often used in jams, preserves, and beverages; are combined in breads and bannocks; and are commonly mixed with other traditional food items. The roots and leaves of berry plants are also sometimes eaten. Moreover, berry harvesting is an important seasonal cultural activity that enhances spiritual, personal, and community well-being for many Inuit (Boulanger-Lapointe et al., 2019). Other species, such as the common puffball (*Lycoperdon perlatum*), are consumed whole (Falandysz et al., 2012).

At high levels, Hg in its various forms can have serious negative health effects on humans. In particular, MeHg is highly toxic to humans, and even in small doses can result in lung and kidney failure; cause damage to the central nervous, digestive, and immune systems; and lead to severe brain damage or death (Petrucci et al., 2011; World Health Organization, 2017). To date, although there are numerous published articles on the impacts of Hg in Arctic wildlife species, literature on the accumulation in edible plant species is very limited, and, to our knowledge, is nonexistent for edible mushroom species of the Canadian Arctic. Therefore, the main objectives of this study were to evaluate the total Hg content in edible plant and fungal species identified by local Inuit residents as well as in co-located soils of the Canadian Arctic and to build on local knowledge of potential contaminated terrestrial plant and fungi species.

2 | MATERIALS AND METHODS

2.1 | Site description

This study was completed during the summers of 2018 and 2019 in Iqaluit, Nunavut, and the surrounding area. Iqaluit, located at 63.7467° N, 68.5170° W, is the capital of Nunavut and is the largest city in the Canadian Eastern Arctic, with a population of 7,740 (Google Earth, 2019; Statistics Canada, 2016). It is part of the Canadian Arctic Archipelago; thus, the landscape is mainly Arctic tundra. It has a desert-like environment with cold conditions, low temperatures (average of −34 °C in the winter and 7.5 °C in the summer), short growing seasons (50–60 d), little precipitation, and nutrient-poor soils. Once the snow cover melts, ponds and creeks form, but the consistency of the permafrost layer and the short grow-

ing seasons allow for only specific vegetative types to thrive (Mallory & Aiken, 2012; UCMP, 2004).

2.2 | Sample collection

In total, 84 samples from 23 plant species, 28 samples from five lichen species, and 157 samples from eight fungal (mushroom) species were collected and assessed for Hg levels (Table 1). These species were identified as edible and naturally occurring by local community members. Additionally, three soil samples were taken from each of the eight sampling locations (Figure 1). These locations were identified by local community members as favored sites with thriving vegetation during the summer months. These locations include Sylvia Grinnell Park; the Road to Nowhere; the Boat Launch; the Old Airport; the Hill; the Nunavut Research Institute (NRI); the Creek; and one location near Iqaluit, the Island ($n = 24$ soil samples total) (Figure 1). “Unknown” fungi species in Table 1 are those species that resemble a species of the Arctic. Proper permits were gathered for the sampling of soil and vegetation, and this research was supported by the community members and the Nunavut Impact Review Board.

Plant and fungi samples were harvested by carefully loosening the soil around the root systems with a trowel and shaking off excess soil. Plants were placed in labeled Whirlpak bags, deposited in a cooler, and transported to the NRI. Select plant samples for which various components were known to be edible and commonly used by the local Inuit were separated by scissors into their corresponding components (flowers, stems and leaves, roots, and seeds [mountain sorrel only]). Samples were cleaned and rinsed using deionized water and left to air dry.

Soil samples were obtained at a depth of 0–10 cm using a trowel. Soil samples were put in labeled Whirlpak bags, placed in a cooler, and transported to the NRI for analysis. Sample preparation and analysis were completed in Iqaluit. Trowels and scissors were washed with methanol after each use. Wet weights of plants, fungi, and soils were recorded, and samples were dried in weigh boats at room temperature (~26–30 °C) in a vented oven for >48 h or until constant weight. Dried samples were weighed, and the plant and fungi samples were ground using a mortar and pestle and placed in individual labeled Whirlpak bags until analysis.

2.3 | Analytical procedures

Approximately 50 mg of sample was weighed into boats (quartz or nickel) and placed in a Milestone DMA-80 direct Hg analyzer to assess for Hg concentrations using cold vapor atomic absorption spectrophotometry at 253.65 nanometers (nm). Calibration curves were made using a fixed

TABLE 1 Common, Latin and Inuktitut names for plant, lichen and fungal species and the individual components sampled in Iqaluit, Nunavut

Common name	Scientific name	Inuktitut name	Plant components sampled
Alpine bistort (<i>n</i> = 3)	<i>Bistorta vivipara</i>	Tursaq	flowers, stem and leaves
Alpine sweetvetch (<i>n</i> = 3)	<i>Hedysarum americanum</i> (Michx.) Britton	unknown	flowers, stem and leaves
Arctic bladder campion (<i>n</i> = 3)	<i>Silene involucrate</i>	Nakasuujaik	flowers, stem and leaves
Arctic cotton grass (<i>n</i> = 3)	<i>Eriophorum scheuchzeri</i>	Pualunnguat	flowers, stem and leaves
Arctic finger lichen (<i>n</i> = 11)	<i>Dactylina arctica</i>	Nirnait	all
Arctic poppy (<i>n</i> = 3)	<i>Papaver radicum</i>	Igutsat Niqingit	flowers, stem and leaves
Arctic thrift (<i>n</i> = 3)	<i>Armeria maritima</i> subsp. <i>sibirica</i>	Immulik	flowers, stem and leaves
Arctic willow (<i>n</i> = 7)	<i>Salix arctica</i>	Uqaujaq	flowers, stem and leaves
Black lichen (<i>n</i> = 3)	unknown sp.	Nirnait	all
Blueberry (<i>n</i> = 4)	<i>Vaccinium caespitosum</i>	Kitgutangirnait	all (berries)
Common dandelion (<i>n</i> = 3)	<i>Taraxacum officinale</i>	Misartaq	flowers, stem and leaves
Common puffball (<i>n</i> = 52)	<i>Lycoperdon perlatum</i>	Pujualuk	all
Crowberry (<i>n</i> = 3)	<i>Empetrum nigrum</i> subsp. <i>Hermaphroditum</i>	Paurnagaqutik	all
Dwarf fireweed (<i>n</i> = 3)	<i>Chamerion latifolium</i>	Paunnat	flowers, stem and leaves, roots
Fairy ring (<i>n</i> = 7)	<i>Marasmius oreades</i>	unknown	cap and stem
Puffball (<i>n</i> = 19)	<i>Calvatia cretacea</i>	Atungaujaik	all
Labrador tea (<i>n</i> = 3)	<i>Rhododendron tomentosum</i> subsp. <i>Decumbens</i>	Qijuktaaqpait	flowers, stem and leaves, roots
Lamp moss (<i>n</i> = 4)	<i>Schistostega pennata</i>	Maniq	all
Lapland lousewort (<i>n</i> = 4)	<i>Pedicularis lapponica</i>	Ujungnaq	flowers, stem and leaves, roots
Large-flowered wintergreen (<i>n</i> = 5)	<i>Pyrola grandiflora</i>	Igutsait niqingit	flowers, stem and leaves
Mica cap (<i>n</i> = 7)	<i>Coprinellus micaceus</i>	unknown	all
Mountain sorrel (<i>n</i> = 4)	<i>Oxyria digyna</i>	Qunguliit	flowers, stem and leaves, roots
Nodding saxifrage (<i>n</i> = 3)	<i>Saxifraga cernua</i>	Nunaraq qupanuap niqinga	flowers, stem and leaves
Orange lichen (<i>n</i> = 3)	<i>Caloplaca marina</i>	Nirnait	all
Prickly saxifrage (<i>n</i> = 6)	<i>Saxifraga tricuspidata</i>	Kakillarnat	flowers, stem and leaves
Rockweed (<i>n</i> = 3)	<i>Fucus distichus</i>	Irkutiit	all
Russula (<i>n</i> = 7)	<i>Russula</i> sp. (unconfirmed)	unknown	cap and stem
Saffron milkcap (<i>n</i> = 5)	<i>Lactarius deliciosus</i> (unconfirmed)	unknown	cap and stem
Saffron webcap (<i>n</i> = 58)	<i>Cortinarius croceus</i> (unconfirmed)	unknown	cap and stem
Sculpted puffball (<i>n</i> = 2)	<i>Calvatia sculpta</i>	Pujualuk	all
Sea sandwort (<i>n</i> = 3)	<i>Honckenya peploides</i>	Maliksuagait	flowers, stem and leaves
Snow lichen (<i>n</i> = 7)	<i>Flavocetraria nivalis</i>	Nirnait	all
Snowbed willow (<i>n</i> = 3)	<i>Salix herbacea</i>	Quarait	all
White mountain heather (<i>n</i> = 4)	<i>Cassiope tetragona</i>	Qijuktaat	all
Whiteworm lichen (<i>n</i> = 3)	<i>Thamnolia vermicularis</i>	Nirnait	all
Yellow oxytrope (<i>n</i> = 3)	<i>Oxytropis maydelliana</i>	Airag	flowers, stem and leaves, roots

concentration of Hg and varying the volume loaded into the quartz boat. This method was developed by Milestone and is based on USEPA Method 7473 (USEPA, 2007). Concentrations are reported in dry weight since several of the plants and fungi species are dried prior to consumption or usage.

The proportion of the total plant mass of Hg within each individual plant component is shown in Figure 2. This was obtained by multiplying the concentration of Hg in each plant component by the weight of that plant component. This gave the mass of Hg (μg) in the plant component. To obtain the



FIGURE 1 The eight sampling locations in Iqaluit, Nunavut. Map obtained and altered from Google Earth (2019)

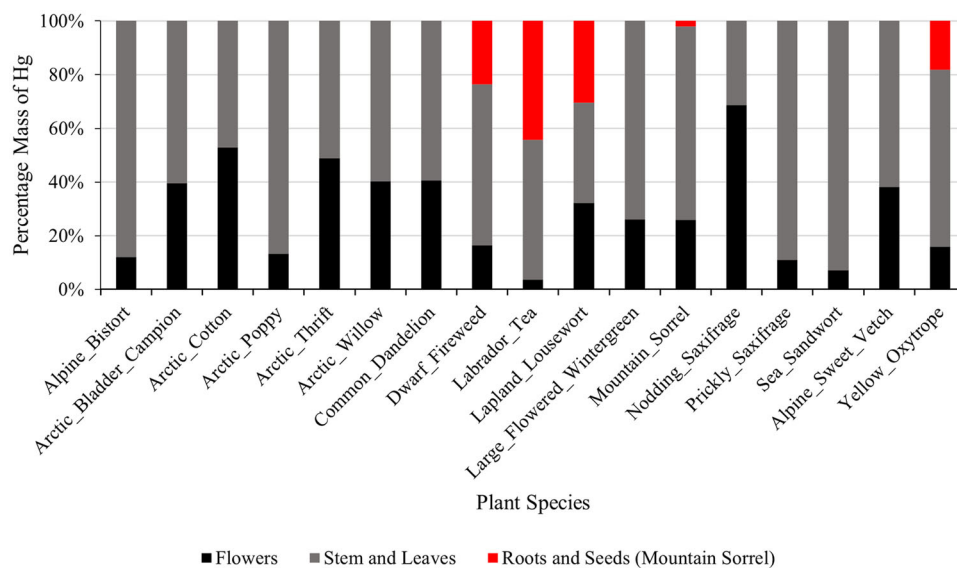


FIGURE 2 Mean mass percentage of Hg from individual plant components

mass of Hg (μg) for the whole plant, the masses of each component were summed. The mass in each component was divided by the sum mass of Hg in the entire plant to get the percentage in each component.

Analysis of MeHg was performed on puffball mushroom samples at the University of Montreal (Quebec, Canada) in the Department of Biological Sciences. Tissues were dried in an oven at 70°C . Reagents used were CH_3HgCl (Alfa Easar Chemicals), methanol, acetic acid, and hydrochloric acid. Sample weights were approximately 0.05 g dry weight.

A quantity of 5 ml of 4 M HNO_3 was added to each sample, and the sample was transferred to a vial containing deionized water (Milli-Q) acetate buffer and $\text{NaB}(\text{Et})_4$. The analysis for MeHg was done with a Tekran 2700 MeHg Auto Analysis System, and the certified reference material TORT-2 (National Research Council Canada) was used for quality assurance.

To determine the extent of the elevated levels of Hg in puffballs in the Arctic, a comparison was also made with samples of puffballs collected from the Kingston, Ontario region to

determine the levels of Hg in puffballs in a non-Arctic and close-by location. Three samples of *Calvatia gigantea* were sampled and tested and had mean Hg concentrations ranging from 3.1 to 7.6 $\mu\text{g g}^{-1}$ Hg dw. These concentrations were similar to the levels found in the Arctic; however, they did not exceed the maximum concentration found in one Arctic puffball sample of 8.2 $\mu\text{g g}^{-1}$ Hg dw.

2.4 | Quality assurance and quality control

For the total Hg analyses, blanks were run with each run and were $\leq 0.003 \mu\text{g g}^{-1}$, assuming an instrumental default mass of 0.1 g. We used 1 $\mu\text{g g}^{-1}$ (20 ng) and 5 $\mu\text{g g}^{-1}$ (200 ng) separate source standard solutions as quality control checks, and were all within 15%. For every 10 samples, a certified reference material and a duplicate were completed. Three certified reference materials were used, with concentrations of Hg as follows: Spinach Leaves SRM 1570a (NIST, 2014) with $30 \pm 6 \text{ ng g}^{-1}$, Tomato Leaves SRM 1573a (NIST, 2018) with $34.1 \pm 1.5 \text{ ng g}^{-1}$, and Lichen BCR 482 (Institute for Reference Materials & Measurements, 2007) with $48 \pm 2 \text{ ng g}^{-1}$. A matrix spike (duplicate sample plus 20 ng Hg ± 6 ng) was completed for every new matrix (soil and plant material) by spiking 20 μl of a 1 $\mu\text{g g}^{-1}$ solution directly onto the unknown sample. Duplicates, blanks, spikes, and matrix spikes were all within the quality control ranges. All duplicates were within 20% of each other, and quality control spikes were within 20% of the expected spiked values. The reliable detection limit of 5 ppb was determined by analyzing eight replicate low level (~ 6 ppb) cabbage reference samples (European Commission, Community Bureau of Reference, Reference Material #679) and calculated as 2 multiplied by the standard deviation multiplied by the *t* statistic (95% confidence limit). The certified reference material TORT-2 was analyzed four times, and values averaged $100 \pm 3\%$ of the certified value for MeHg.

2.5 | Statistical analysis

Concentrations obtained from the Hg analyzer were initially separated by plant components, which were then combined using corresponding masses to determine the concentration of the total plant. Statistical analysis was performed using the free statistical software R x 64 3.4.3. Concentrations in samples that were measured below the detection limit ($< 0.005 \mu\text{g g}^{-1}$ for Hg) were recorded as half of the detection limit and included in the analysis. All statistical tests were conducted at a .05 level of significance. Descriptive statistics including mean, standard deviation, and median soil Hg concentrations for each sampling location were first calculated. Due to the non-normal nature of the data, a Kruskal–Wallis test was

TABLE 2 Mean soil concentration, corresponding SD, and median concentration of mercury in samples collected from the eight sampling locations in Iqaluit, Nunavut and the surrounding area ($n = 3$ for each sampling location)

Sampling location	Mean Hg concentration $\mu\text{g g}^{-1}$	SD	Median Hg concentration $\mu\text{g g}^{-1}$
Airport	0.047	0.064	0.011
Boat Launch	0.063	0.051	0.070
Creek	0.017	0.004	0.020
Hill	0.042	0.026	0.031
Island	0.042	0.011	0.046
Nunavut Research Institute	0.056	0.030	0.044
Road to Nowhere	0.036	0.033	0.023
Sylvia Grinnell Park	0.16	0.22	0.041

conducted to determine if there was a significant difference in soil Hg concentrations between sampling locations. Based on findings indicating that soil Hg concentrations were similar between all sampling locations, sampling location was not investigated in further statistical analyses, and data were combined from different locations.

Descriptive statistics including mean, standard deviation, and median total Hg concentration of each plant and fungi species were obtained. Kruskal–Wallis tests were used to determine if Hg concentrations differed between (a) plant and (b) fungi species. Based on preliminary findings, a nonparametric unpaired *t* test (Mann–Whitney U test) was used to determine if fungi classified as “puffballs” had higher Hg concentrations than “non-puffballs.” Species, sampling locations, and percent moisture of each sample can be found in the Supplemental Table 1.

3 | RESULTS AND DISCUSSION

3.1 | Soil Hg concentration by sampling locations

Individual soil Hg concentrations ranged from 0.01 $\mu\text{g g}^{-1}$ (Boat Launch) to 0.41 $\mu\text{g g}^{-1}$ (Sylvia Grinnell Park). Means, standard deviations, and median soil Hg concentration for each sampling location can be found in Table 2. There was no significant difference between soil Hg concentrations from the eight sampling locations (Kruskal–Wallis $\chi^2 = 4.6$; $df = 7$; $p = .71$).

Results are consistent with Hg concentrations in those reported in other Northern or Arctic (surface permafrost, tundra, and wetland) soils from Alaska, Cornwallis Island, Northwest Territories, and Norway, which had mean values ranging

from 0.046 to 0.111 $\mu\text{g g}^{-1}$ and median values up to 0.114 $\mu\text{g g}^{-1}$ (Gamberg et al., 2015; Halbach et al., 2017; Leitch, 2006; Loseto et al., 2004; Obrist et al., 2017; Olson et al., 2018). The nearest relevant locations (Cornwallis Island, Northwest Territories) ranged from 0.01 to 0.30 $\mu\text{g g}^{-1}$ in individual samples, and the reported range in concentrations of Northern soils in the literature and the present study are both higher than examples in temperate and tropical soils (0.020–0.050 $\mu\text{g g}^{-1}$) (Obrist et al., 2017).

The Canadian soil quality guidelines recommended by the Canadian Council of Ministers of the Environment suggest a maximum concentration of 6.6 $\mu\text{g g}^{-1}$ Hg for human health in residential/parkland and agricultural land use (CCME, 1999). In this study, all soil samples were below the recommended guideline; the mean soil Hg concentration from all locations was 0.019 $\mu\text{g g}^{-1}$ ($n = 24$). Although this study had a larger range of concentrations, the mean value was similar to the lower end found by Obrist et al. (2017) from temperate locations and is consistent with other previous studies (Leitch, 2006; Loseto et al., 2004; Obrist et al., 2017). Most of the sampling locations were near commercial or residential buildings and were disturbed by human activity, which may have reduced deposition of Hg due to the removal of top layers of soil and increased runoff from roads, sidewalks, and housing on these locations. Levels in areas far from anthropogenic activities such as Sylvia Grinnell Park and the Boat Launch were slightly higher, with mean concentrations of 0.16 ± 0.22 and 0.063 ± 0.051 $\mu\text{g g}^{-1}$ Hg, respectively.

3.2 | Plant and lichen Hg concentrations by species

Of the 110 plant samples obtained, total Hg concentrations in the individual plant components ranged from below the detection limit (<0.005 $\mu\text{g g}^{-1}$ in Lapland lousewort) to 0.71 $\mu\text{g g}^{-1}$ (large-flowered wintergreen). Means, standard deviations, and median Hg concentration for each plant and lichen species can be found in Table 3. There was a significant difference in total Hg concentration between certain plant species (Kruskal–Wallis $\chi^2 = 64.6$; $df = 27$; $p < .01$), where nodding saxifrage (*Saxifraga cernua* L.) had the lowest median concentration (0.005 $\mu\text{g g}^{-1}$), and yellow oxytrope [*Oxytropis campestris* (L.) DC] had the highest (0.19 $\mu\text{g g}^{-1}$).

3.3 | Plant Hg concentration by plant component

Yellow oxytrope, the plant with the highest median Hg concentration, had the highest mean levels of Hg in the stem and leaf components, with 66% of the mass of Hg found in those

components. A similar finding was seen for the common dandelion (*Taraxacum officinale* F.H. Wigg.) and prickly saxifrage (*Saxifraga tricuspidata* Rottb.) (59 and 90% of Hg in the stem and leaves, respectively). Most of the Hg within the plant species where parts were separated had accumulated in the stem and leaf components of the plants compared with other components, such as the roots and flowers. Because there are no trees or tall shrubs in the Arctic to absorb the atmospheric Hg, researchers have presumed that a large percentage of the Hg in the plant shoots derives from the atmosphere (Dombaiová, 2005; Tomiyasu et al., 2005). It is hypothesized that GEM enters plant leaves by the same pathway as carbon dioxide, through the various stoma on the underside of plant leaves. Once absorbed, Hg may remain in the Hg(0) form or transform to Hg(II) and MeHg forms, and all these forms bind to the plant tissues and accumulate before being deposited back into the soil after plant death and decomposition (Gamberg et al., 2015; Obrist et al., 2017). In general, levels of MeHg in plants are minimal compared with the other forms of Hg. A study by Bailey et al. (2002) conducted in Alaska found that levels of MeHg accounted for 0.1–2.7% of the total Hg found within willow and alder species. Similar trends were observed by Dombaiová (2005) in deciduous and coniferous trees from nine locations in Slovakia.

Most research on metal accumulation in terrestrial Arctic vegetation focuses on species found in northern European countries (Falandysz et al., 2012; Klos et al., 2012), with only three studies looking at fungi and terrestrial plants of the Canadian Arctic (Chiarenzelli et al., 2001; Choy et al., 2010; Gamberg, 2009). Mercury concentrations in various leaf and twig samples (including cotton grass, willow, and saxifrage species similar to those in the present study) from Nunavut and Yukon (maximum concentration of 0.057 $\mu\text{g g}^{-1}$) were similar to those in the present study. Lichen concentrations in these studies ranged from 0.072 to 0.26 $\mu\text{g g}^{-1}$ (Chiarenzelli et al., 2001; Choy et al., 2010; Gamberg, 2009), which was slightly higher but similar to the lichen concentrations in the present study, which ranged from 0.034 to 0.11 $\mu\text{g g}^{-1}$. These studies concluded that lichens had higher levels of total Hg compared with the plant species analyzed. Lichens survive the winter months (and may even be active and productive [Kappen, 1993]), which may promote the accumulation of additional contaminants during spring snow melt. However, Hg concentrations in the lichen species analyzed in this study were not as high as those found in some of the plant species. This could be a result of the absent root system in lichens, which limits uptake, allowing only Hg to accumulate from the atmosphere (Gamberg et al., 2015). The highest median Hg concentration of all lichen samples was 0.11 $\mu\text{g g}^{-1}$ Hg dw in orange lichen, which was similar to levels seen by Choy et al. (2010) but slightly lower than levels in plant species in this study, such as yellow oxytrope and common dandelion.

TABLE 3 Mean concentration, SD, median concentration, and sample size collected from combined plant samples of 28 plant species from Iqaluit, Nunavut and the surrounding area

Species	Sample size (<i>n</i>)	Mean Hg concentration μg g ⁻¹ dry wt.	SD	Median Hg concentration μg g ⁻¹ dry wt.
Alpine bistort	3	0.094	0.12	0.025
Alpine sweet vetch	3	0.018	0.002	0.017
Arctic bladder campion	3	0.12	0.19	0.022
Arctic cotton grass	3	0.021	0.022	0.008
Arctic finger lichen	11	0.035	0.007	0.034
Arctic poppy	3	0.059	0.037	0.053
Arctic thrift	3	0.086	0.11	0.028
Arctic willow	7	0.021	0.016	0.016
Black lichen	3	0.094	0.044	0.072
Blueberry	4	0.016	0.007	0.015
Common dandelion	3	0.13	0.13	0.027
Crowberry	3	0.010	0.006	0.010
Dwarf fireweed	3	0.044	0.015	0.039
Labrador tea	3	0.037	0.013	0.044
Lamp moss	4	0.061	0.044	0.043
Lapland lousewort	4	0.007	0.030	0.006
Large flowered wintergreen	5	0.27	0.27	0.11
Mountain sorrel	4	0.091	0.06	0.073
Nodding saxifrage	3	0.010	0.008	0.005
Orange lichen	3	0.11	0.02	0.11
Prickly saxifrage	6	0.085	0.035	0.099
Rockweed	3	0.013	0.005	0.012
Sea sandwort	3	0.059	0.008	0.061
Snow lichen	7	0.059	0.022	0.053
Snowbed willow	3	0.039	0.021	0.039
White mountain heather	4	0.040	0.021	0.038
Whiteworm lichen	3	0.069	0.051	0.097
Yellow oxytrope	3	0.15	0.10	0.19

3.4 | Fungi Hg concentration by species

Of the 157 mushroom samples obtained, individual total Hg concentrations ranged from 0.007 μg g⁻¹ (*Cortinarius croceus*) to 8.2 μg g⁻¹ (*L. perlatum*). Means, standard deviations, and median Hg concentrations for each fungi species can be found in Table 4. There was a significant difference in total Hg concentrations between fungi species (Kruskal–Wallis $\chi^2 = 99.8$; $df = 7$; $p < .01$), where *C. croceus* had the lowest median concentration (0.084 μg g⁻¹), and *L. perlatum* had the highest (1.6 μg g⁻¹).

3.5 | Fungi Hg concentration by type (puffball vs. non-puffball)

There was a significant difference in Hg concentration between puffball species and non-puffball species (W-

statistic = 311; $p < .01$), where the median Hg concentration in puffball species (1.4 μg g⁻¹) was approximately 12 times higher than non-puffball species (0.12 μg g⁻¹) (Figure 3).

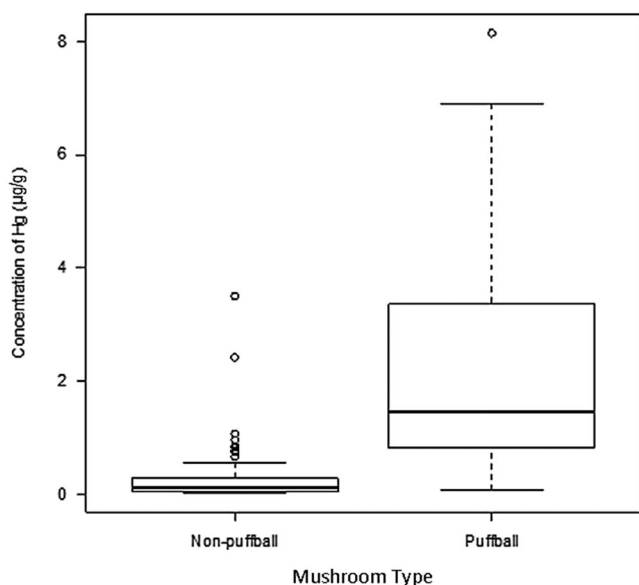
Uptake, translocation, and accumulation of Hg and other metals through the mycelium into the aboveground components of mushrooms is generally species-, genera-, and family-dependent, which makes it difficult to compare different species and locations (Melgar et al., 2009). Fungi are composed predominantly of mycelium, which grows in the soil and on litter and/or organic matter. The mycelium is usually extremely long-lived and can spread over a large area. Therefore, mycelium may be exposed to high contamination and if accumulated (i.e., stays in the organism rather than fluxes in and out) could translate into high concentrations, potentially making its way into the fruiting body of the fungi.

The mycelium grows fruiting bodies usually on an annual basis, and the fruiting bodies contain spores. During the

TABLE 4 Mean Hg concentration, SD, median Hg concentration, and sample size collected from eight fungi species sampled in Iqaluit, Nunavut and the surrounding area

Species	Type	Sample size (<i>n</i>)	Mean Hg concentration $\mu\text{g g}^{-1}$ dry wt.	SD	Median Hg concentration $\mu\text{g g}^{-1}$ dry wt.
<i>Calvatia cretacea</i>	puffball	19	2.0	1.5	1.3
<i>Calvatia sculpta</i>	puffball	2	0.64	0.057	0.64
<i>Coprinellus micaceus</i>	non-puffball	7	0.17	0.16	0.097
<i>Cortinarius croceus</i> (unconfirmed)	non-puffball	58	0.23	0.39	0.084
<i>Lactarius deliciosus</i> (unconfirmed)	non-puffball	5	1.1	1.4	0.74
<i>Lycoperdon perlatum</i>	puffball	52	2.4	2.0	1.6
<i>Marasmius oreades</i>	non-puffball	7	0.28	0.18	0.34
<i>Russula</i> sp. (unconfirmed)	non-puffball	7	0.21	0.098	0.20

Note: dw, dry weight.

**FIGURE 3** Boxplot of mercury concentrations found in fungi species considered puffballs ($n = 73$) vs. non-puffballs ($n = 84$)

lifetime of the fruiting body (from days to weeks, depending on the species), spores can be spread for reproductive purposes. The spores may be spread by rain or wind or drop to the existing soil or organic matter and may germinate. Literature on Hg accumulation in fungi is very limited, but it is predicted that Hg is predominantly absorbed through the soil and enters fungi and plants by the same process as micronutrients. The low Hg concentrations of soil samples analyzed in this study, and the high concentrations in puffball species suggest that Hg accumulates from alternative sources, including direct absorption from the atmosphere. However, according to Falandysz et al. (2016), mushrooms have no known mechanisms to take in Hg from atmospheric sources, and the mycelial network absorb a majority of the chemical elements

from the substrate. On the other hand, puffball biomass has been shown puffballs to be potential biosorbents of Hg due to their large affinity for metal accumulation (Sari et al., 2012). It is unknown how Hg is taken up and accumulated within these puffball species, and further investigation is required.

In two studies, several samples of the common puffball (*L. perlatum*) in Poland had large masses and were able to accumulate and store large amounts of Hg, ranging from 0.57 to 4.5 $\mu\text{g g}^{-1}$ Hg dw (Falandysz et al., 2012; Falandysz et al., 2003). Both studies showed similar levels of Hg in the species *L. perlatum* compared with the samples analyzed in this study, where a median value of 1.6 $\mu\text{g g}^{-1}$ Hg dw was observed. There is minimal research on Hg levels in fungi of the Canadian Arctic, with only one study reporting Hg concentrations ranging from 0.15 to 0.44 $\mu\text{g g}^{-1}$ dw in six unidentified fungal samples collected in the Yukon (Choy et al., 2010). Our fungi medians for *C. cretacea*, *C. sculpta*, *L. perlatum* (puffball species), and *L. deliciosus* (unconfirmed non-puffball) were higher than the concentrations reported in Choy et al. (2010).

When young, puffballs have a white, fleshy interior, with one of the highest protein contents in fungal species and are readily consumed by Indigenous peoples of Canada (Arnason et al., 1981). Once puffballs mature into readiness for spore release, the interior turns into a brown/green powder, which is inedible but not toxic to humans (Falandysz et al., 2012). All puffballs analyzed in this study were collected in the young, edible stage and in the similar stages to fungi obtained in the literature stated above.

3.6 | Hg speciation

Mercury's toxicity varies according to its form. In its methylated form (MeHg and Me_2Hg), samples are more toxic and can result in serious health effects for humans (World Health

Organization, 2017). Samples of puffball mushroom species were analyzed for MeHg to better assess their toxicity. *Lycoperdon perlatum* had a mean MeHg concentration of $0.085 \pm 0.074 \mu\text{g g}^{-1}$ MeHg dw ($n = 3$), *C. cretacea* had a concentration of $0.032 \pm 0.088 \mu\text{g g}^{-1}$ MeHg dw ($n = 2$), and a single *C. sculpta* had a concentration of $0.013 \mu\text{g g}^{-1}$ MeHg dw. These MeHg concentrations represented 1.6–2.3% of the total Hg (mean, $2.2 \pm 0.5\%$) in the samples. Based on these findings, most of the Hg within the highest contaminated samples is not MeHg. Additional speciation analysis of Hg in these samples would determine how inorganic Hg is bound within these organisms, which may help with understanding the high concentrations. Although minimal amounts of MeHg were found, high levels in inorganic forms are still a concern to humans and wildlife species consuming the fungi, and the tendency for MeHg to biomagnify represents a vector for MeHg into higher trophic level wildlife in the terrestrial environment (Evers, 2018).

3.7 | Food safety implications

The Canadian sampling location and potential consumption of samples analyzed in the present study dictate that Canadian food standards are applicable for food safety considerations. For Hg, no Canadian food standards are available for any food item other than fish. According to Health Canada (2018), the maximum allowable levels of Hg (total Hg) in various types of fish products sold in Canada is $0.5 \mu\text{g g}^{-1}$ fresh weight (fw). In the absence of any other guidelines, this fish guideline is used for the present study. The guideline is judged to be sufficiently protective for plant and fungi samples because it is likely that the consumption rates of the plants and fungi sampled are lower than those of fish. Additionally, the fish guideline is based on the majority of Hg in a sample consisting of MeHg, with less toxic inorganic Hg predominating in the plant and fungi samples, as shown by this study and by Bailey et al. (2002) and Dombaiová (2005). Because there are discrepancies in moisture content between fish and the samples in the present study (plants are drier than fish and fungi are wetter), both dw and fw concentrations are compared. No median plant levels of Hg were above $0.5 \mu\text{g g}^{-1}$ dw and therefore not above the $0.5 \mu\text{g g}^{-1}$ fw guideline (due to more dilution with water weight). For fungi, on a dry weight basis, median levels of Hg in *L. perlatum* ($1.6 \mu\text{g g}^{-1}$ dw), *C. cretacea* ($1.3 \mu\text{g g}^{-1}$ dw), *C. sculpta* ($0.64 \mu\text{g g}^{-1}$ dw), and *L. deliciosus* ($0.74 \mu\text{g g}^{-1}$ dw) were above the $0.5 \mu\text{g g}^{-1}$ fw guideline. On a fresh weight basis, only the mean value of Hg in *L. perlatum* of $0.79 \mu\text{g g}^{-1}$ fw and $0.56 \mu\text{g g}^{-1}$ in *C. cretacea* are above the $0.5 \mu\text{g g}^{-1}$ fw guideline. As mentioned, the minimal amounts of MeHg in these fungi suggest that the comparison to the fish guideline is overly conserva-

tive, although a detailed human health risk assessment may be a prudent next step.

4 | CONCLUSION

This study aimed to evaluate the total Hg levels in soils as well as edible plant, lichen, and fungi species of Iqaluit, Nunavut, and the surrounding area. Across the eight sampling locations, soil concentrations of Hg did not statistically differ. Of all plant and fungi samples, the puffball fungi accumulated the highest concentrations of Hg. Based on the high levels of Hg, puffballs were also assessed for MeHg, where three species showed mean concentrations ranging from 0.013 to $0.085 \mu\text{g g}^{-1}$ MeHg dw. Overall, this study provides insight into the levels of Hg found in various Canadian Arctic plant and fungi species as well as in the soils and is the first to focus on Hg levels in several plants used by local Inuit. Further, this study contributes information on the processes of Hg accumulation in Arctic floral species and provides awareness for the need to mitigate Hg levels in Arctic environments.

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AUTHOR CONTRIBUTIONS

Ryan Bergin: Conceptualization; Data curation; Formal analysis; Investigation; Methodology; Project administration; Resources; Software; Validation; Visualization; Writing-original draft. Allison Rutter: Conceptualization; Funding acquisition; Investigation; Project administration; Supervision; Writing-review & editing. Iris Koch: Conceptualization; Investigation; Writing-review & editing. Jamal Shirley: Investigation; Methodology; Writing-review & editing. Barbara Zeeb: Conceptualization; Project administration; Supervision; Writing-review & editing.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

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