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Review of Ecological Mercury and Arsenic Bioaccumulation within Historical Gold Mining Districts of Nova Scotia

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ABSTRACT

Gold mining in Nova Scotia dates back to the mid-1800s. Historical industrial practices generated over 3,000,000 tonnes of finely-ground mine wastes (tailings) which were deposited into lakes, streams, wetlands and low-lying areas close to the mill sites. These legacy tailings typically contain high concentrations of mercury (Hg) and arsenic (As) and continue to impact downstream environments to this day. The objective of this review is to critically examine and summarize existing knowledge on the transfer and bioaccumulation of Hg and As in aquatic and terrestrial organisms exposed to legacy gold mine tailings in Nova Scotia. This review reveals that as of mid-2019, 24 previous studies have been completed on this subject. Several of these studies were based on small sample sizes, or had other limitations, such as missing identification of species. Despite these limitations, the data in these publications clearly indicate that both Hg and As from abandoned gold mine sites in Nova Scotia are bioaccumulating in plants, fungi, freshwater and terrestrial invertebrates, marine mollusks, amphibians, fish and mammals. In many cases, concentrations of Hg and As in tissue exceed Canadian Council of Ministers of the Environment (CCME) safety guideline values for wildlife consumption. No studies were found examining tailings-related Hg or As accumulation in lichen, reptiles, or large mammals. This review concludes that further research on bioaccumulation and biomagnification of tailings-related Hg and As is needed to understand the overall impact of historical tailings on the aquatic and terrestrial ecosystems and species of Nova Scotia. More detailed studies are vital for guiding risk-management decisions and future land-use practices for these contaminated sites.

INTRODUCTION

Gold mining has been an important part of Nova Scotia's history and culture since the mid-1800s. There are 64 formal gold mining districts containing hundreds of individual mines, stretching across mainland Nova Scotia (Bates, 1987; Art Gallery of Nova Scotia, 2013). The initial discovery of bedrock gold mineralization occurred near Mooseland in 1858 (Malcolm, 1929). In the decades that followed, the gold industry became an integral part of Nova Scotia's economy and played a major role in the development of many small mining towns. Between 1861 and the mid-1940s, there were three distinct gold rushes resulting in the production of an estimated 1.2 million troy ounces (approximately 37,000 kg) of gold (Bates, 1987). This is likely a conservative estimate, as historical mining companies commonly underestimated the amount of recovered gold to avoid paying royalties to the government (Art Gallery of Nova Scotia, 2013).

During this era of gold mining, most auriferous (gold containing) ore in Nova Scotia was crushed to sand- or silt-sized material using stamp mills or ball mills, then the gold was extracted using Hg amalgamation techniques (Blakeman, 1978). Fresh water from nearby aquatic ecosystems was used to wash the crushed ore over Hg-coated copper plates. Free gold grains formed an amalgam with the Hg, which was scraped from the plates and heated to boil off Hg, thus leaving the remaining gold. It has been estimated that one ounce of Hg was used for every ounce of gold recovered, although higher estimates of Hg per ounce of gold have been reported (Parsons et al., 2012). Beginning in the 1890s, gravity separation, roasting, chlorination, and cyanidation were also added to the milling circuit at some mines to recover gold from sulphide minerals and older tailings, but Hg amalgamation remained an essential part of most mills until the 1940s.

Residual sand- to silt-sized mill waste (also called tailings) was commonly deposited into nearby lakes, streams or wetland ecosystems, after the gold was extracted. There are currently an estimated 3,000,000 tonnes of historical gold mine tailings throughout the province (Parsons et al., 2012). Although most Nova Scotian gold mining districts are now long-abandoned, a legacy of significant environmental contamination at the historical stamp mill sites and tailings disposal areas remain today (Parsons et al. 2012; Drage, 2015). The Hg recovery process during the late 1800s to early 1900s was inefficient, and an estimated 10-25% of the Hg used was lost to tailings or to the atmosphere during this time (Eaton, 1978; Parsons & Percival, 2005).

Along with Hg, tailings also typically have elevated As concentrations (Parsons et al. 2012). Most gold is hosted by quartz veins within meta-sandstones and slate of the Cambro-Ordovician Meguma Supergroup, which makes up most of the southern mainland of Nova Scotia. Arsenopyrite (FeAsS) is the most commonly occurring sulphide mineral in these auriferous veins and surrounding host rocks and contains 46 wt.% As by mass (Kontak & Jackson, 1999). When arsenopyrite in tailings and other mine waste is exposed to the atmosphere, it slowly weathers and oxidizes, releasing As into the surrounding environment (Lengke et al., 2009).

Mercury is not an essential element required for biological functioning, and many forms of Hg can be highly toxic, including dimethylmercury, methylmercury (MeHg) and elemental mercury (Hg^0). Hg^0 was the form used for gold amalgamation at historical gold mines in Nova Scotia, and is still used today in artisanal gold mines in some countries. It is a silvery metal, liquid at room temperature and highly volatile. When Hg^0 enters into an aquatic environment, either from mining wastes or atmospheric deposition, it may oxidize, forming inorganic Hg (Hg^{II}), which has a much higher solubility in water (O'Driscoll et al., 2005). When Hg^{II} is present in acidic environments with anaerobic conditions, high microbial activity, and high levels of organic carbon (i.e. many wetland conditions), it may undergo methylation by microorganisms such as sulphate-reducing bacteria (SRB) and ferric-reducing bacteria (FeRB), which can produce organic Hg compounds, including MeHg (O'Driscoll et al., 2005).

Methylmercury is especially toxic to living organisms, highly bioavailable, and lipophilic, allowing it to pass through membranes, including the blood-brain barrier and the placental barrier (Park & Zheng, 2012). In this way, MeHg can reach vital organs and accumulate in the brain (CCME, 2003; Edmonds et al., 2010; Doe et al., 2017). Additionally, MeHg is eliminated from the body very slowly, making it more likely to bioaccumulate and biomagnify in higher predators (O'Driscoll et al., 2005). Studies have shown that MeHg accounts for the majority of total Hg (THg) accumulation in aquatic organisms (Lavoie et al., 2013).

Arsenic is a metalloid with highly complex chemistry and over fifty naturally occurring species, each with varying toxicities to humans and other organisms (Cullen & Reimer, 1989; Francesconi & Kuehnelt, 2004). The toxicity of As depends greatly on its speciation, bioavailability, concentration and the detoxification mechanisms of exposed organisms (Cullen & Reimer, 1989). In general, contrary to Hg, inorganic forms of As (IAs) are more toxic than organic forms, as they are also proven carcinogens (Mass et al., 2001; Ng, 2005). It is these inorganic species, including arsenite (As^{III}) and arsenate (As^{V}), which often enter into freshwater systems following weathering of mine tailings (Campbell and Nordstrom, 2014). Microbial reactions may also release As from the solid to aqueous phase, increasing the mobility of As in freshwater environments (Oremland & Stolz, 2003).

Arsenic has been shown to accumulate in biota through their contact with, and ingestion of, contaminated water, sediment, soil and/or organic matter, but despite numerous studies, there remains much uncertainty around the biomagnification potential of As (Rahman & Hasegawa, 2012). Within living organisms, As is mainly found in organic forms, such as arsenobetaine (often found in marine organisms) and arsenocholine, both of which are relatively non-toxic (Cullen & Reimer, 1989). However, more toxic inorganic species have been found to bioaccumulate in both terrestrial and aquatic organisms, including in a number of species within Nova Scotia including plants (Koch et al., 2007; Saunders et al., 2011), invertebrates (Chapman et al., 2016; Moriarty et al., 2009), shellfish (Koch, 2007; Whaley-Martin et al., 2012, 2013; Walker and Grant, 2015), amphibians (Moriarty et al., 2013; Saunders et al., 2010, 2011), and mammals

(Moriarty et al., 2011; Saunders et al., 2010, 2011) among others (Rahman & Hasegawa, 2012).

The first environmental studies of gold mine tailings in Nova Scotia took place in the late 1970s, after a resident living near a historical gold district (Waverley) was diagnosed with chronic As intoxication following exposure to contaminated well water (Hindmarsh et al., 1977). Several studies over the subsequent four decades have shown that tailings from historical gold mines have high concentrations of both As and Hg and, at some sites, have contaminated downstream environments (e.g. Eaton, 1978; Eaton & Clair, 1985; Wong et al., 1999; Koch et al., 2007; Parsons et al., 2012). From 2003 to 2012, detailed studies by Natural Resources Canada, several universities, and other government departments helped to characterize the environmental and human health hazards associated with 14 of these historical gold mine districts and provided guidance for site management and remediation (Parsons et al., 2012; Drage, 2015). The CCME has provided Canadian As and Hg guidelines for the protection of aquatic life and environmental and human health. Maximum limits have been set for Hg in sediment (0.486 ppm dw, freshwater, 0.700 ppm dw marine), soil (6.6 ppm, residential) and water (0.026 ppb freshwater, 0.016 ppb marine), as well as for As in sediment (17 ppm dw freshwater, 41.6 ppm dw marine), soil (12 ppm) and water (5 ppb freshwater, 12.5 ppb marine) (CCME, 2003, CCME, 2017). However, there are no tissue residue guidelines for the protection of wildlife set for either Hg, MeHg or As to this point.

Parsons et al. (2012) summarize results from a multi-disciplinary investigation of the dispersion, speciation and fate of metal(loid)s in terrestrial and shallow marine environments surrounding 14 of the 64 historical gold mine districts in Nova Scotia. Mercury and As analysis of 482 tailings and sediment samples from these sites showed that As ranged from 10 to 312,000 mg/kg (median = 2250 mg/kg) with 99% of samples exceeding both soil and sediment guidelines, while Hg ranged from <5 μg/kg to 350,000 μg/kg (median = 1640 μg/kg), with 20% exceeding soil guidelines, and 71% exceeding sediment guidelines (Parsons et al., 2012; CCME, 2017). Early results from this and similar studies led to the formation of a Provincial-Federal Historic Gold Mines Advisory Committee (HGMAC) in 2005, which evaluated the ecological and human health risks associated with these gold mines in more detail, and issued warnings to help reduce human exposure to tailings at some sites (Drage, 2015; Nova Scotia Environment, 2017). The potential ecosystem and human health risks at most sites are still not well understood, and many gold mining districts remain unstudied.

The objective of this review is to critically examine and summarize existing articles, publications and reports with information on bioaccumulation of Hg and As in organisms, both aquatic and terrestrial, exposed to historical gold mining waste in Nova Scotia. This includes any living organisms (excluding humans) that could be adversely affected by environmental contamination resulting from historical gold mine tailings. Until this point there has been no systematic review of the bioaccumulation of Hg and/or As in gold mining or tailings-affected sites in Nova Scotia. Future research needs on this topic have also been identified.

METHODS

Documents and publications containing information about legacy gold mine contaminants and biological impacts were located using multiple sources and techniques, including online databases (Web of Science and Google Scholar) and government databases held within the Nova Scotia Department of Natural Resources, Environment and Climate Change Canada, and the Geological Survey of Canada. Reviewing the bibliographies and literature-cited sections of prior reports also led to other documents. Finally, scientists and managers who work on this topic in academia, government and the private sector were directly consulted, and many grey-literature reports and documents only available in hard-copy format were discovered in this way. These reports were digitized to PDF files and uploaded to our internal library for future reference.

At the time of writing, collected studies were produced from 1978 to 2017, with 7 between 1978 and 1989, 2 from the 1990s, 2 from the 2000s, and 12 from 2010 to present. These 23 studies include data from 18 historical gold mining districts and 14 associated reference sites (Table 1, Figure 1). We found information for several broad taxonomic groupings including: (1) freshwater, terrestrial and marine plants; (2) fungi; (3) freshwater and terrestrial invertebrates; (4) marine shellfish; (5) amphibians; (6) fish; and (7) mammals. The review below follows those same broad categories, with results discussed chronologically within each section. Values for [Hg] and [As] in organisms and relevant information (e.g. sample size, type of organism, location) were extracted from tables and text in all papers. If the authors used a reference site (e.g. non-impacted site with no gold mine tailings) to compare with the contaminated site data, we included information on those datapoints as well. We did not need to extract data from graphs in this review. Some reports included speciation data (e.g. inorganic arsenic or MeHg), but the majority reported only total As (TAs) and Hg (THg) concentrations. All Hg and As units were converted to ppm, and described in either dry weight (dw) or wet weight (ww).

A number of studies had limitations such as small sample sizes, or lacked specific identification of species with only common names provided. When possible, the scientific names were interpreted based on the context and description within the reports. Some authors had a broad approach to sampling to maximize the number of environmental matrices. In some studies, only average values or the minimum-maximum values were reported. Longitude and latitude were not always reported, especially in older documents, so data was organized by associated district, mine and waterbody where possible.

The largest gold districts in Nova Scotia often encompass several mines and tailings areas, named after mining companies that operated in the district at a specific point in time. For example, the Montague District includes both the main Montague Gold Mine site and several separately named mines and ore processing sites. There is a reporting bias within this literature review, with the largest datasets and number of studies associated with the Montague, Upper Seal Harbour (USH) and Lower Seal Harbour (LSH) districts (Table 1, Figures 3, 4).

It should be noted that our literature review found no studies examining tailings-related Hg or As accumulation in lichen, reptiles, birds, or large mammals. Although there are other contaminants of concern associated with historic gold mine wastes (e.g. nickel, lead, antimony), this review is limited to Hg and As, as these elements present the greatest potential risk to ecosystem and human health at gold mine sites in Nova Scotia (Parsons et al., 2012; Drage, 2015). Results of this literature review are summarized in a map depicting each study site, and the broad taxonomic groups studied at each location (Figure 1). Studies were also categorized by district to highlight the most studied locations (Table 1), and by taxonomic groups, to show the most studied species and habitats (Supplemental Information, Table 2).

REVIEW

Algae & Plants

Plants are the most commonly researched taxonomic group at legacy gold mine tailings sites in Nova Scotia. Ten studies document [Hg] and/or [As] in plants from tailings-impacted sites (Figure 1). These studies identified over 50 different plant species, to varying levels of taxonomic resolution. The majority of the plant-related publications pertained to terrestrial plant species (*n*=35 species), followed by wetland or freshwater aquatic species (*n*=17 species), with only one publication discussing marine flora (*n*=1 seaweed species; Koch et al., 2007).

The earliest report, Eaton (1978), conducted broad surveys of three gold mine districts (Montague, Mount Uniacke and Oldham) and collected ferns, grasses, mosses and rooted aquatic vegetation during the summer and early autumn of 1977. Plant samples in this study were characterized broadly (e.g., “grass”, “moss”). Mercury concentrations (ww) ranged from 0.01 to 1.05 ppm at Montague, 0.012 to 1.8 ppm at Mount Uniacke, and 0.0004 to 5.8 ppm at Oldham. Overall, aquatic vegetation had higher [Hg] than terrestrial plants, with filamentous and encrusting algae samples from Oldham demonstrating especially elevated [Hg] (up to 5.06 ppm). Although there was little evidence of a correlation between Hg in vegetation and in sediment/soil, the strongest correlation (r^2=0.24) was found at Oldham. No reference data were provided with this study. Terrestrial and shoreline plants (*n*=8 species) at Montague District were again assessed by Dale and Freedman (1982) in a detailed plant survey at various sites along the main tailings flats and in the cross-cutting Mitchell Brook. Average [As] in plants from tailings sites ranged from 11 to 834 ppm dw (median = 101 ppm), compared to 0.5 to 6 ppm (median = 2 ppm) in reference plants from Fleming Park, NS.

Aquatic and semi-aquatic plants (*n*=7 species) at the Oldham District were analyzed by Lane et al. (1988) for [Hg] and [As] concentrations in both the roots and shoots of the plants growing in or near tailing material. Average [Hg] ranged from 0.18 to 16.3 ppm dw (shoots) and 0.47 to 6.11 ppm dw (roots), while average [As] ranged from 321 to 4260 ppm dw (shoots) and from 2650 to 6340 ppm dw (roots) (Lane et al., 1988). Overall, both [Hg] and [As] were highest in root samples, with [As] ranging from 4.4 to 17 times higher than shoot concentrations (Lane et al., 1988). Reference data was presented for As only, ranging from 2 to 13 ppm dw, with concentrations up to 300 times lower that [As] found in plants at Oldham District (Lane et al., 1988). In a subsequent report, Lane et al. (1989) conducted further analysis on these stored plant samples from Oldham (*n*=32 species, *n*= 598 samples). They found that [Hg] ranged from <0.01 to 8.86 ppm dw, while [As] ranged widely from 2 to 10,000 ppm dw (Lane et al., 1989). Plant roots, again, had higher [Hg] and [As] than plant shoots in all plant samples where a comparison was conducted. No reference site data were provided with this study but [As], again, far exceeded the reference data in the Lane et al. (1988) study.

Marine flora were studied only once, by Koch et al. (2007). Seaweed (*Fucus sp*.) collected from a tailings-impacted marine embayment downstream of the Lower Seal Harbour (LSH) District had TAs ranging from 27 to 43 ppm ww, with InAs ranging from 9.4 to 13.2 ppm. This was elevated compared to reference seaweed, which contained TAs ranging from 6 to 10 ppm ww and InAs from 0.75 to 1.0 ppm. Bioaccessible As accounted for 63-81% in seaweed samples overall, and did not differ significantly between organisms at contaminated and reference sites. Authors noted that as the seaweed is attached to rocks rather than growing in contaminated sediment, the As accumulation is likely a result of uptake from the surrounding water (Koch et al., 2007).

Data from Saunders et al. (2011) demonstrated that terrestrial plants from the Upper (USH) and Lower (LSH) Seal Harbour Districts also have elevated [TAs]. Terrestrial plants at USH had mean TAs of 25 ppm (8.8 ppm As(III), 10 As(V)) and plants at LSH had mean TAs of 8.8 ppm (2.4 As(III), 1.6 As(V)), while reference plants had mean TAs of 0.14 ppm (<0.04 As(III), <0.04 As(V)).

Edible plants were studied only once by Koch et al. (2013), who assessed both TAs and As bioavailability in edible berries, among other country foods (see fungi and mammal sections), growing at LSH. They found TAs ranged from 8 to 21 ppm ww in blueberries at the tailing site (*n*=4), and from 0.059 to 0.16 ppm ww in blueberries, blackberries and raspberries from uncontaminated sites (*n*=3). Toxic InAs accounted for a large proportion of the bioavailable fraction in these samples, and authors noted this may be a potential human health risk if berries are indeed being consumed (Koch et al., 2013).

Fungi

Edible fungi species were also included in the Koch et al. (2013) study of tailings-related contaminants in country foods. Two fungi species (*Laccaria laccata* and *Suillus luteus*) were collected from tailings sites within the LSH District. These authors found that *Suillus luteus* had 0.50 ± 0.03 ppm ww TAs (*n*=1, duplicate extraction), while *Laccaria laccata* contained 46 ppm ww (*n*=1). Both species contained predominantly dimethylarsinic acid, and *Laccaria laccata* also contained a significant amount of trimethylarsine oxide (Koch et al., 2013). However, as only one sample was collected for each species, the authors concluded more data were needed before drawing conclusions on As speciation in these species at tailings sites.

Freshwater and Terrestrial Invertebrates

Invertebrates from freshwater and terrestrial habitats were discussed in five studies, including an unpublished B.Sc. honours thesis. The majority of these of the studies were conducted at the Montague District, with some data from the USH and LSH districts (Figure 1). Two of these evaluated [Hg] (Eaton, 1978; Robinson, 2016) while three evaluated [As] (Brooks et al., 1982; Moriarty et al., 2009; Button et al., 2012). A variety of terrestrial invertebrate species (*n*=11) and aquatic freshwater species (or life stages) (*n*=5) were described within the studies, with 14 different species or taxonomic groups identified in total.

Water striders (genus *Gerriae*, *n*=3) collected by Eaton (1978) from Mitchell Brook within the Montague District had [Hg] ranging from 0.13 to 0.25 ppm dw. Brooks et al. (1982) also sampled aquatic invertebrates from along the tailings-impacted area of Mitchell Brook, and found that composite samples of caddisflies (genus *Trichoptera*) and mayflies (genus *Ephemeroptera*) had [As] ranging from 0.002 to 0.059 ppm dw. No reference data were included in either of these studies.

Freshwater invertebrates from Mitchell Brook were again sampled by Robinson (2016) at a tailings-impacted wetland along the brook nicknamed “Old Stamp Mill” (Robinson, 2016, unpublished). Their study, although unpublished, indicated elevated [Hg] in dragonfly larvae (suborder Zygoptera), damselfly larvae (suborder Anisoptera), and aquatic spiders (genus *Dolomedes*). Organisms from the Old Stamp Mill wetland had

[Hg] up to 2.0 ppm dw, while reference invertebrates from an upstream, uncontaminated site along Mitchell Brook had mean [Hg] ranging from 0.17 to 0.24 ppm dw.

Terrestrial invertebrates at historical tailings sites have been described in two publications (Moriarty et al., 2009; Button et al., 2012). Moriarty et al. (2009) assessed [As] amongst a variety of taxonomic groups (including spiders, grasshoppers, ants, flies and earthworms) collected from the Montague, USH and LSH districts. They found that organisms from all categories had elevated TAs compared to those at reference sites, including moths which had up to 22 ppm dw As at Montague, compared to 0.13 ppm dw at a reference site along East Brook (Moriarty et al., 2009). Earthworms (*L. castaneous* and *D. rubidus)* at the LSH District were also found to have exceptionally elevated [As] by Button et al. (2012). Concentrations of TAs were up to 2200 ppm dw in organisms from LSH, compared to 4.3 ppm dw at the reference site at New Harbour (Button et al., 2012). This marked the highest [As] reported to date in earthworms.

Marine Shellfish

Five studies, to date, examine tailings-related [As] in marine shellfish at tailings-impacted coastal sites, two of which also include [Hg]. The first study published on shellfish was conducted by Koch et al. (2007), who found that soft-shelled clams (*Mya arenaria*) from Seal Harbour (downstream of the LSH District) had elevated TAs, ranging from 218 to 228 ppm ww, compared to reference clams (unidentified species, purchased from grocery store) with TAs ranged from 7.0 to 7.9 ppm ww. Blue mussels (*Mytilus edulis*) were later tested along a gradient of tailings contamination in Seal Harbour by Whaley-Martin et al.

(2012). Results showed that composite mussel samples (*n*=10-15) had average TAs of 60-109 ppm dw, with InAs of 15-33 ppm ww, while reference organisms from Coddles Harbour and a local grocery store, had TAs of 16-34 ppm dw and InAs of 0.2-7.92 ppm dw. Authors suggested that this concentration of IAs may present a risk to any higher trophic organisms (including humans) consuming the mussels (Whaley-Martin et al., 2012).

Periwinkles (*Littorina littorea*) from Seal Harbour also had exceptionally elevated TAs (Whaley-Martin et al. 2013). Average TAs concentrations in organisms from LSH ranged from 400 to 840 ppm dw, with [InAs] from 290-588 ppm dw, while reference periwinkles from Coddles Harbour had 56 ppm ww TAs, with 6.6 ppm InAs. These concentrations of As were attributed, in part, to the feeding mechanism of the periwinkles, which scrape seaweed off rocks and other substrate, and therefore are in close contact with tailings-contaminated sediment (Whaley-Martin et al., 2013). The [InAs] accumulated in these periwinkles represents, to date, the highest concentrations reported in any marine organisms. Again they were noted to be a potential risk to marine consumers (e.g. crab and bird species) and to humans harvesting edible periwinkles.

Walker & Grant (2015) also analyzed blue mussels, along with American lobsters (*Homarus americanus*) from Isaacs Harbour (Isaacs Harbour District). Historically, tailings were discharged directly into deeper waters in Isaacs Harbor, with minimal impact on the intertidal zone (Parsons et al., 2008). In addition, Isaacs Harbour does not receive contaminated freshwater runoff from mine tailings, which is a major source of exposure for shellfish in Seal Harbour (Milligan and Law, 2013). Accordingly, the mussels from these sites demonstrated lower [As] than those tested by Whaley-Martin et al (2012, 2013). Mussels had TAs of 1.3-2.0 ppm ww, and [Hg] of 0.02-0.05 ppm ww, while lobster hepatopancreas tissue had TAs of 5.0-10.0 ppm ww and THg of 0.06-0.12 ppm ww. These results demonstrated significantly elevated [Hg], but not [As], found in organisms from the contaminated Isaacs Harbour site, compared to the reference Country Harbour site. The authors noted that while no mussels exceeded CFIA guidelines, a number of lobster tissue samples exceeded CFIA Hg guidelines for fish and fish products (CFIA, 2011) at both reference and contaminated sites.

Despite much study of coastal locations near the USH and LSH gold districts, there were no publications for Hg or As in species at any other coastal tailings sites until 2017, when Doe et al. (2017), released a paper on blue mussels (*Mytilus edulis*) and soft shelled clams (*Mya arenaria*) collected from eight different historical gold districts. The highest [As] was found in organisms from Seal Harbour (up to 309 ppm ww), however, all 8 sites demonstrated elevated [As] when compared with reference organisms from New Harbour. Mollusks collected near the Goldenville, Gold River and Wine Harbour districts also had elevated [As] compared to the reference site, demonstrating that other coastal tailings sites are also affecting nearby marine environments.

Amphibians

In their 1978 report, Eaton (1978) included [Hg] for frogs (*n*=16) and toads (*n*=1) of unidentified species, living within the Oldham, Mount Uniacke and Montague gold districts. The amphibians had [Hg] ranging from 0.1 to 0.45 ppm ww at Oldham (*n*=4), from 0.16 to 0.32 ppm ww at Mount Uniacke (*n*=7), and from 0.03 to 0.06 ppm ww at Montague (*n*=6), however, no reference data were provided (Eaton, 1978). Only one other publication reported on amphibians. Moriarty et al. (2013) reported on [As] in the leg tissue of green frogs (*Rana clamitans, n*=11) and an eastern American toad (*Bufo americanus*, *n*=1) collected from the USH District. Amphibians at USH had elevated [TAs], ranging from 1.6 to 4.4 ppm ww (2.7 ± 1.2 ppm ww average), while reference animals had average TAs of 0.23 ± 1.2 ppm ww TAs (Moriarty et al., 2013).

Fish

The effects of historical tailings on fish species have been described in six publications, mentioning 12 predominantly freshwater species (Eaton, 1978; Brooks et al., 1982; Dale and Freedman, 1982; Eaton and Clair, 1985; Tetford, 1999; LeBlanc & Halfyard, 2010).

Eaton (1978) included a number of eel (*n*=14), bass (*n*=1), perch (*n*=5) and trout (*n*=5), collected from the Montague and Oldham districts, with [Hg] of 0.12-0.75 ppm ww (Eaton, 1978). No reference data were included, but this initial study demonstrated a number of fish from both sites that surpassed the current Hg CFIA safety guideline for edible fish tissue of 0.5 ppm ww (CFIA, 2011).

An exposure experiment was later conducted by Dale and Freedman (1982), by placing 10 banded killifish (*Fundulus diaphanous*) originally caught from a non-contaminated site (Fleming Park, NS) in contained cages for four weeks in the tailings affected Mitchell

Brook (Montague District). The study site was located in an area previously determined to have the highest water [As] in that stream (Brooks et al., 1982). Results showed significant As accumulation in fish caged in Mitchell Brook, (0.639 ppm ww) compared to fish caged at a reference site (0.446 ppm ww), by the end of the experiment. Two native banded killifish were also accidentally caught in the Mitchell Brook cages during the experiment, and were found to have [As] of 4.02 and 4.77 ppm ww; surpassing the current TAs CFIA guidelines of 3.5 ppm ww for edible fish tissue (CFIA, 2018).

Eaton and Clair (1985) also found elevated Hg in fish caught from four tailings-affected lakes within the Waverley District. They collected brook trout (*n*=5), white perch *n*=12), small mouth bass (*n*=3), and white suckers (*n*=7), and found that Hg ranged from 0.11 to 2.00 ppm ww. Although the study lacked reference data, 15 of 27 fish samples (from a variety of species) exceeded the current recommended CFIA consumption safety guideline of 0.50 ppm ww Hg (CFIA, 2018).

White perch (*Morone americana*) from the Caribou District were studied by Tetford (1999), who examined [Hg] in fish from Long Lake, which is partially filled with gold mine tailings. Average [Hg] in fish from Long Lake (*n*=9) was 0.62 ppm dw, roughly 3 times that of fish from the uncontaminated Angevine Lake (*n*=12) which had average [Hg] of 0.23 ppm dw. Fish from Long Lake also had shorter average fork lengths (138.78 mm vs. 147.58 mm reference), and lower average weights (33.67 g vs. 50.33 g reference), with Long Lake fish weighing up to 40% less than fish of equal length from the reference site (Tetford, 1999). The author noted that both sites were small, land locked, oligotrophic lakes of the same region, and therefore hypothesized that differences in [Hg], size and mean fork length were potentially attributed to the influence of the tailings material (Tetford, 1999, unpublished).

In the most extensive and recent fish study in Nova Scotia, LeBlanc and Halfyard (2010) provide an in-depth analysis of contaminants in 11 fish species from 13 different tailings-contaminated lakes, compared to fish from six reference lakes (n=300 fish total). Fish from tailings-affected lakes had average [Hg] of 0.62 ppm ww (11 species, n=172), which was slightly higher, though not statistically significant, from the average [Hg] in fish from non-contaminated lakes of 0.57 ppm ww (7 species, n=45) (LeBlanc and Halfyard, 2010). Fish [Hg] was positively correlated with diet, and in some species, with length and weight. There was also some evidence that fish from tailings-contaminated lakes had higher [TAs], after a comparison of lakes using an analysis of co-variance (ANCOVA); however, most fish did not exceed the CFIA TAs guideline of 3.5 ppm ww (LeBlanc and Halfyard, 2010).

Small Mammals

Four mammal species (from five publications) have been studied for bioaccumulation of tailings-specific Hg and/or As in Nova Scotia. Mammals include meadow voles (*Microtus pennsylvanicus*), shrews (*Sorex cinereus*), hares, and mice of unspecified species (Eaton, 1978; Saunders et al., 2010, 2011; Koch et al., 2013). A number of small rodents (mice of unspecified species) were first included in the Eaton (1978) survey. Animals from

Oldham, Mount Uniacke and Montague districts had [Hg] of 0.01-0.064 ppm ww (*n*=4), with the highest concentration found in a mouse at Mount Uniacke (Eaton, 1978).

Small rodents from the Montague District were again assessed by Saunders et al. (2010), who conducted a study on meadow voles (*Microtus pennsylvanicus*) living at or near tailings flats. Vole [As] tissues ranged from 2.1 to 6.2 ppm ww (*n*=10), compared to 0.25 to 0.53 ppm ww (*n*=10) at a nearby reference site off Montague Road (Saunders et al., 2010). Saunders et al. (2011) published similar results in voles from USH and LSH districts, where the animals had [TAs] of 0.52-2.5 ppm ww, compared to 0.19-0.48 ppm ww at a reference site. The animals were known to feed mainly on terrestrial invertebrates, which were previously shown to be highly contaminated at the Montague, USH, and LSH gold districts (Moriarty 2009; Button et al., 2012). Accordingly, prey items and stomach contents of the voles were analyzed, and elevated [As] were found; 7.6 ppm ww in animals from Montague, and 4.2 ppm ww in animals from the Seal Harbour area, compared to 0.44 and 0.48 ppm ww in animals from reference sites, respectively (Saunders et al., 2010, 2011). Similarly, Moriarty et al. (2011) showed that shrews (*S. cinereus*) from USH also had elevated mean As when compared to a control site (0.28 ppm ww vs. 0.1 ppm ww).

Finally, hunted game species living near tailings sites were assessed in only one study. Koch et al. (2013) in their survey of foraged and hunted ‘country foods’, found that TAs in a hare caught near tailings at Seal Harbour (LSH District) had 0.021 ppm ww (*n*=1), compared with animals from reference sites, which had TAs of 0.44 and 0.007 ppm ww (*n*=2). Bioaccessible As in hare tissue was predominatly InAs, although some samples with lower TAs had InAs below detection limits. Authors noted that the tissue was analyzed uncooked, but that cooking would likely occur prior to human consumption, which may change As speciation (Koch et al., 2013).

Non-Tailings Related Studies

In addition to studies specifically linked to tailings-sourced Hg and As, there have also been a number of other recent studies demonstrating elevated [Hg] in mammals and bird species within Nova Scotia.

In a 2015 study by Little et al., elevated [Hg] was found in fur samples of little brown bat collected from Nova Scotia when compared with Canadian background levels (Environment and Climate Change Canada, 2016). The study included fur samples from animals at many locations across Atlantic Canada, but only two bats were collected specifically within historical gold mining districts. Fur samples from these two animals had [Hg] of 15.7 ppm ww in a specimen caught within the Rawdon District, and 5.68 ppm ww in one from the Chezzetcook District. In stable isotope analysis, bats from Atlantic Canada were found to have mean δ13C consistent with freshwater invertebrates in the area. However, given the small sample size, we are not able to assess whether the elevated Hg was connected with historical tailings deposits. In general, the bats sampled from Nova Scotia had the highest [Hg] in all of Atlantic Canada (Little et al., 2015). Background [Hg] in little brown bats from Ontario and Quebec had [Hg] of 5.2 and 2.4 ppm ww, respectively (Environment and Climate Change Canada, 2016). The effects of tailings-related contaminants may be even more important to understand now, after the rapid decline of bat populations following the outbreak of white-nose-syndrome and their inclusion on the endangered species list in Nova Scotia in 2013 (NSDNR, 2013).

Spencer et al. (2011) examined Hg accumulation in the brains of river otters (*Lontra Canadensis*) in Nova Scotia. Brain tissues from otters (*n*=66) showed [Hg] ranging from 0.3 to 18 ppm ww. Although this was not directly linked to tailings-related sources, some of the animals were collected from areas with a high density of abandoned mines (Spencer et al., 2011). On a national scale, Nova Scotia is one of the only provinces where [Hg] in the livers of mink and otter had been shown to surpass the threshold for neurochemical effects (15 to 51 ppm dw) (Environment and Climate Change Canada, 2016).

No studies were found for tailings-related contaminants in birds, however, a number of studies have demonstrated elevated Hg in birds across Nova Scotia when compared with national background concentrations. Edmonds et al. (2010) conducted a study on [Hg] in blood and feather samples of the rusty blackbird (*Euphagus carolinus nigrans*), an endangered species within Nova Scotia (Environment & Climate Change Canada, 2015), and a wetland obligate bird, whose feeding and breeding range spans the whole of Nova Scotia. Birds collected from Acadian forests demonstrated mean blood [Hg] of 1.06 ppm ww, and a geometric mean of 8.26 ppm ww in feather samples (*n*=59) (Edmonds et al., 2010). The study included specimens from New Brunswick, New Hampshire, Maine and Vermont, but the highest overall blood [Hg] of 3.42 ppm ww, was found in a bird from

Nova Scotia (Edmonds et al., 2010). The [Hg] in this animal far surpassed minimum levels know to result in adverse health effects in birds (Evers et al. 2008). In addition, MeHg accounted for approximately 98% of the THg in blood samples, and 97% in feather samples (Edmonds et al., 2010). Although the elevated mercury levels found in this species were not directly linked to historic tailings sources, historic gold mine tailings were listed as a possible cause of the elevated THg and MeHg levels found in the birds (Edmonds et al., 2010).

Finally, a number of studies have shown elevated [Hg] and [MeHg] in invertebrates, fish and loons within Kejimkujik National Park, NS (Burgess et al., 2008; Buckland-Nicks et al., 2013). Burgess et al. (2008) demonstrated that as fish and loon blood [Hg] increased, productivity decreased in the bird. Although elevated [Hg] in this area has not been directly linked to historical tailings deposits, there are a number of gold mine districts surrounding the park area.

DISCUSSION

In total, 23 studies completed since 1978 were reviewed examining [Hg] and [As] in living organisms collected at, or near, historical gold mine tailings sites in Nova Scotia. Many of these sites have elevated [Hg] and [As] in water, sediment and soil, often at concentrations surpassing Canadian guidelines for the protection of aquatic life and environmental health (Eaton, 1978; Brooks et al., 1982; Wong et al., 1999; Parsons et al., 2012). Of the studies reviewed, 19 gold mining districts were mentioned in total. Lower Seal Harbour was the most-studied district (*n*=10) resulting in publications on fungi, plants, invertebrates, marine shellfish, amphibians and fish. Seal Harbour, located downstream of the Lower Seal Harbour mine, also remains the only site to be closed to food harvesting, after early findings on tailings-related mollusk contamination led to a ban on shellfish harvesting in 2005 (Environment Canada, 2005). Montague was the second most studied site (*n*=7), followed by Upper Seal Harbour (*n*=4).

Reference data were included in 15 studies, and in all cases, organisms from tailings-impacted sites exceeded [Hg] and/or [As] found in reference organisms (Dale and Freedman, 1982; Lane et al., 1988; Tetford, 1999; Koch et al., 2007; Moriarty et al., 2009, 2011, 2013; LeBlanc and Halfyard., 2010; Saunders et al., 2010, 2011; Button et al., 2012; Whaley-Martin et al., 2012, 2013; Koch et al., 2013; Robinson, 2016; Doe et al., 2017). Although there is currently no maximum acceptable limit for Hg or As for the protection of wildlife consumers, there are numerous examples of species collected near tailings sites with [Hg] known to result in adverse health effects in wildlife (Evers et al. 2008; Edmonds et al., 2010; Spencer et al., 2011 Environment and Climate Change Canada, 2016). The data summarized in these publications indicate that Hg and As are bioaccumulating in plants, invertebrates, amphibians, fish and mammals living near many of these historical gold mine districts. However, more research is needed to fully understand the effects of gold mine tailings on both species and ecosystem health.

Plants are the most studied taxonomic group at historical tailings sites, with nine different studies on tailings-related contaminant bioaccumulation, and data collected on plants across ten different historical districts (Figure 1). There is strong evidence that plants growing in tailings, soil or sediment at gold mine sites have elevated [As]. Maximum [TAs] in plants from the Oldham (6340 ppm dw), Montague (834 ppm dw) and Upper and Lower Seal Harbour (34 ppm ww) districts far exceed maximum [TAs] found in plants from associated reference sites (13 ppm dw, 6 ppm dw, 0.93 ppm ww, respectively). Studies on Hg are sparser, and baseline [Hg] for plant tissue in Nova Scotia is lacking and needed to contextualize plant [Hg] findings from the Montague, Mount Uniacke and Oldham districts. There is evidence that coastal tailings sites are influencing marine flora, and resulting in contaminant accumulation (Whaley-Martin et al., 2012, 2013; Doe et al., 2017).

Terrestrial and aquatic invertebrates from tailing sites have also been shown to have elevated [Hg] and [As]. However, studies are currently limited to three districts: Montague, USH and LSH. Aquatic invertebrates from the "Old Stamp Mill" wetland at Montague demonstrate up to 8 times higher [Hg] than associated reference organisms (Robinson, 2016, unpublished). Terrestrial invertebrates at tailings sites had [As] over 160 times (Montague) and 300 times (USH and LSH) higher than reference organisms (Moriarty et al., 2009; Button et al., 2012). Evidence of contaminant transfer to small mammals feeding on terrestrial invertebrates was demonstrated at these sites as well, as discussed in the mammals section (Saunders et al., 2010, 2011; Moriarty et al., 2011, 2013). Invertebrates are common ecological receptors of contaminants as they live in direct contact with soil, water and/or sediment and have an especially high surface area-to-mass ratio. Furthermore, emergent (i.e. hatching) insects with aquatic life stages have been shown to transfer contaminants such as Hg and As to terrestrial ecosystems (Mogren et al., 2013; Tweedy et al., 2013). In this way, they can act as biovectors of contaminants to higher trophic levels and to surrounding ecosystems. (Eaton & Clair, 1985; Koch et al., 2007; LeBlanc and Halfyard., 2010; Doe et al., 2017).

Marine invertebrates have also been affected by tailings disposal in the coastal zone. Many shellfish (e.g. molluscs, crustaceans, and echinoderms) are commonly harvested for consumption along the coast of Nova Scotia by residents (Fisheries & Oceans Canada, 2018). Unfortunately, a number of popular shellfish harvesting sites along the eastern shore of the province are also known areas of historical gold mine tailing contamination. These sites include: Seal Harbour (LSH District), Isaacs Harbour (Isaacs Harbour District), Wine Harbour (Wine Harbour District), Harrigan Cove (Harrigan Cove District) and Gegogan Harbour (Goldenville District) among others (Parsons et al., 2012; Doe et al., 2017). The Canadian Food Inspection Agency (CFIA) currently has no set guidelines for Hg or As limits in shellfish, but does provide guidelines for fish tissue, limiting concentrations to 0.5 ppm ww Hg, and 3.5 ppm ww As (CFIA, 2011, 2018). Maximum limits for shellfish have also been set for [IAs] in New Zealand (2 ppm ww), Hong Kong (10 ppm ww), and Australia (1 ppm ww) (Edmonds & Francesconi, 1993). The Seal Harbour clams, mussels and periwinkles have [InAs] far surpassing these concentrations (Koch et al., 2007; Whaley-Martin et al., 2012, 2013; Walker and Grant, 2015; Doe et al., 2017).

Prior to 2005, shellfish from Seal Harbour were harvested for food by residents in the area, but preliminary data from Environment Canada (summarized in Doe et al. 2017), led to a ban on harvesting bivalve mollusks at Seal Harbour in 2005 (Contaminated Fisheries Prohibition Order No. STN-20050007; Environment Canada, 2005). At present, this remains the only location in Nova Scotia where harvesting of wild foods has been restricted due to contamination from gold mine waste (Environment Canada, 2007; Parsons et al., 2012). Additional studies on marine mollusks should be carried out to better understand contamination at other less-studied coastal tailings sites and to assess the risk these mollusks may present to higher-order consumers (Whaley-Martin et al., 2012, 2013; Walker & Grant, 2015).

Amphibians are often used as bioindicators of environmental conditions as a result of both their physiology (thin skin, cutaneous respiration) and their lifecycle, which includes stages in both freshwater and terrestrial environments (Moriarty et al., 2013). Although studies on amphibians near Nova Scotia gold mine sites are limited to only two reports, the data presented by Moriarty et al. (2013) show elevated [As] in amphibians from the USH District when compared to reference organisms.

Fish are economically important for Nova Scotia, and consumed by both humans and wildlife. Sportfishing in Nova Scotia generates annual revenues of over $58 million, with nearly 67,000 licensed anglers, the majority of whom are permanent residents of the province (Nova Scotia Dept. of Fisheries & Aquaculture, 2018). Overall, fish are the second most studied category of species. However, fish can be challenging bioindicators for understanding overall site-specific contamination, as many species in Nova Scotia are highly mobile and migratory, possibly traveling outside the spatial bounds of the contaminated sites to feed. As a result of this behavior, fish species may also act as important biovectors of contaminants beyond the tailings sites themselves. The reports gathered on fish species here show that there have been numerous cases of [Hg] in fish surpassing the CCME and CFIA guidelines at the Montague District (Eaton, 1978; Brooks et al., 1982; Dale & Freedman, 1982), Oldham District (Eaton, 1978), Waverley District (Eaton and Clair, 1985), and Caribou District (Tetford, 1999). However, LeBlanc and Halfyard (2010) also demonstrated a lack of overall elevated average [Hg] in fish from tailings-impacted lakes across Nova Scotia, when compared with reference sites. More research is needed to fully understand the role fish may play in the transfer of Hg and As from tailings-impacted sites to wildlife and human consumers.

California, similar to Nova Scotia, has an extensive history of historical gold mining dating back to the 1800s. As a result of publications demonstrating elevated Hg in both fish and invertebrates associated with historical and active gold mine sites (Slotton et al. 1995; Klasing & Brodberg, 2003, 2004), California issued fish consumption advisories for over 20 waterbodies affected by gold mining activities (California Dept. of Conservation, 2002). The state has also produced public documents warning the public about waterbodies that are known as tailings-related Hg “hot spots” (OEHHA, 2018). In 1997, the Department of Conservation in California also created the federally funded Abandoned Mine Lands Program (AMLP) to address the health, safety and contamination concerns associated with historical mine land (California Dept. of Conservation, 2002).

Nova Scotia does provide human fish consumption guidelines with the goal of limiting Hg exposure in the yearly Sportfishing Anglers Handbook (Nova Scotia Dept. of Fisheries & Aquaculture, 2018). However, while site-specific warnings are in place for lakes with elevated PCBs, there are not yet site-specific warnings for lakes or waterbodies connected to historical gold mine tailing deposits, which may be Hg and/or As "hotspots" (Nova Scotia Dept. of Fisheries & Aquaculture, 2018). This is despite the fact that [Hg] surpassing the provincial and federal safety guidelines have already been found in fish from numerous districts (see Fish section above).

Finally, small mammals and rodents living near tailings sites demonstrate elevated [Hg] and [As] at the Montague, USH, and LSH districts, but have not been studied to date in other locations. Only one study was found which included species commonly hunted for consumption in Nova Scotia (Koch et al., 2013), and no studies on larger mammals (i.e. deer, moose) have been conducted.

This literature review demonstrates that there are still significant gaps in our understanding of ecosystem impacts from historical gold mine tailings in Nova Scotia, and the transfer of Hg and As from tailings into local food webs. A lack of data for many of the gold mining districts and species living within them makes it difficult to assess the degree to which tailings-exposed ecosystems are being affected compared to non-contaminated watersheds and terrestrial habitats within the province. More studies are especially important in areas where activities like wild berry picking, hunting game, shellfish harvesting, or fishing may be taking place. Detailed studies of species- and ecosystem-level contaminant concentrations and effects are essential for developing successful management and remediation practices for these tailings sites.

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REFERENCES

Art Gallery of Nova Scotia (2013) Gold: A Nova Scotia Treasure (virtual exhibit available online at: http://www.novascotiagold.ca). (August 6, 2018).

Bates, J.L.E. (1987) Gold in Nova Scotia. Nova Scotia Department of Natural Resources, Mineral Resources Branch, Information Series ME 13, 48. Available online at: https://novascotia.ca/natr/meb/pdf/is13.asp

Blakeman, W.B. (1978) An inventory of the known gold mines in Canada which practiced mercury amalgamation, 1855–1975. Unpublished report for Environment Canada, 144 p.

Brooks, R. R., Fergusson, J. E., Holzbecher, J., Ryan, D. E., Zhang, H. F., Dale, J. M., & Freedman, B. (1982) Pollution by arsenic in a gold-mining district in Nova Scotia. *Environmental Pollution Series B, Chemical and Physical*, 4(2), 109–117.

Brooks, R.R., Holzbecher, J., Ryan, D.E. (1981) Horsetails (*Equisetum*) as indirect indicators of gold mineralization. *Journal of Geochemical Exploration*, 16, 21-26.

Burgess, N. M., & Meyer, M. W. (2008). Methylmercury exposure associated with reduced productivity in common loons. *Ecotoxicology*, *17*(2), 83-91.

Buckland-Nicks, A., Hillier, K. N., Avery, T. S., & O'Driscoll, N. J. (2014). Mercury bioaccumulation in dragonflies (Odonata: *Anisoptera*): Examination of life stages and body regions. *Environmental Toxicology and Chemistry*, *33*(9), 2047–2054.

Button, M., Koch, I., & Reimer, K. J. (2012) Arsenic resistance and cycling in earthworms residing at a former gold mine in Canada. *Environmental Pollution*, 169, 74–80.

California Department of Conservation (2002) California's Abandoned Mines: A Report on the Magnitude and Scope of the Issue in the State. Retrieved online at http://www.conservation.ca.gov/dmr/abandoned_mine_lands/AML_Report/ (July 2 2018)

Campbell, K. M., and Nordstrom, D. K. (2014) Arsenic speciation and sorption in natural environments. *In:* R.J. Bowell, C.N. Alpers, H.E. Jamieson, D.K. Nordstrom, & J. Majzlan (Eds.), Arsenic: Environmental Geochemistry, Mineralogy, and Microbiology. Reviews in Mineralogy and Geochemistry 79: 185-216.

Canadian Council of Ministers of the Environment (CCME) (2003) Inorganic mercury and MeHg. In: Canadian environmental quality guidelines, 1999. Canadian Council of Ministers of the Environment, Winnipeg.

Canadian Council of Ministers of the Environment (CCME) (2017) Canadian Environmental Quality Guidelines and Summary Table. Retrieved online at: http://ceqg-rcqe.ccme.ca/ (August 1, 2018).

Canadian Food Inspection Agency (CFIA) (2011) Canadian Guidelines for Chemical Contaminants and Toxins in Fish and Fish Products. Available online at: http://www.inspection.gc.ca/english/fssa/fispoi/man/samnem/app3e.shtml.

Canadian Food Inspection Agency (CFIA) (2018) List of Contaminants and other Adulterating Substances in Foods. Available online at: https://www.canada.ca/en/health-canada/services/food-nutrition/food-safety/chemical-contaminants/contaminants-adulterating-substances-foods.html (March 24, 2019).

Chapman, E. E. V., Robinson, J., Berry, J., & Campbell, L. M. (2016) Can a Low-Dose Selenium (Se) Additive Reduce Environmental Risks of Mercury (Hg) and Arsenic (As) in Old Gold Mine Tailings? *Water, Air, & Soil Pollution*, *227*(6), 216.

Cullen, W. R., & Reimer, K. J. (1989) Arsenic speciation in the environment. *Chemical Reviews*, *89*(4), 713–764.

Dale, J. M., & Freedman, B. (1982) Arsenic pollution associated with tailings at an abandoned gold mine in Halifax County, Nova Scotia. *Proceedings of the Nova Scotian Institute of Science*, 32, 337-349.

Doe, K., Mroz, R., Tay, K.-L., Burley, J., Teh, S., & Chen, S. (2017) Biological effects of gold mine tailings on the intertidal marine environment in Nova Scotia, Canada. *Marine Pollution Bulletin*, 114, 64-76.

Drage, J. (2015) Review of the Environmental Impacts of Historic Gold Mine Tailings in Nova Scotia. Nova Scotia Dept. of Natural Resources, Open File Report ME 2015-004.

Eaton, P. (1978) Investigation of Mercury Related Environmental Effects Associated with Abandoned Amalgamation Operations in Nova Scotia. Environmental Protection Service (EPS) Atlantic Region, Environment Canada Report 234, 58 p.

Eaton, P. & Clair, T.A. (1985) Fish Contamination in Shubenacadie Headwater Lakes. *In:* A. Mudroch and T.A. Clair (Eds.), The Impact of Past Mining Activities on the Shubenacadie River Headwaters Ecosystem. *Environment Canada / N.S. Dept. of the Environment Report, IWD-AR-WQB-85-81, 195 p.*

Edmonds, S. T., Evers, D. C., Cristol, D. A., Mettke-Hofmann, C., Powell, L. L., McGann, A. J., … O'Driscoll, N. J. (2010) Geographic and Seasonal Variation in Mercury Exposure of the Declining Rusty Blackbird - Variación Geográfica y Estacional en la Exposición al Mercurio de la Especie en Disminución Euphagus carolinus. *The Condor*, *112*(4), 789–799.

Edmonds, J. S., & Francesconi, K. A. (1993) Arsenic in seafoods: human health aspects and regulations. *Marine Pollution Bulletin*, *26*(12), 665-674.

Environment Canada (2005) Toxic Chemicals Update Newsletter. Retrieved from http://atlanticweb1.ns.ec.gc.ca/epb/newsletters/toxchem/Default.asp?lang=En&*n*=1A776D33-1 (June 29, 2018).

Environment Canada (2007) A legacy of pollutants in Nova Scotia: Assessing risks, taking action. S&T into Action Series, Catalogue No. En164-15/1-2007E-PDF.

Environment and Climate Change Canada (2015) Committee on the Status of Endangered Wildlife in Canada. https://www.canada.ca/en/environment-climate-change/services/committee-status-endangered-wildlife.html. (April 13, 2018).

Environment and Climate Change Canada (2016) Canadian Mercury Science Assessment Report. Government of Canada, Catalog Number En84-130/3-2016E-PDF, 793 p. Available online at http://publications.gc.ca/site/eng/9.810484/publication.html (April 13, 2018).

Evers, D. C., Savoy, L. J., DeSorbo, C. R., Yates, D. E., Hanson, W., Taylor, K. M., Fair, J. (2008) Adverse effects from environmental mercury loads on breeding common loons. *Ecotoxicology*, *17*(2), 69–81.

Francesconi, K. A., & Kuehnelt, D. (2004) Determination of arsenic species: A critical review of methods and applications, 2000–2003, *Analyst*, 129(5), 373–395.

Fisheries and Oceans Canada (2018) Shellfish harvesting openings and closures. Retrieved from http://www.dfo-mpo.gc.ca/shellfish-mollusques/closures-fermetures-eng.htm (July 1 2018).

Hindmarsh, J.T., McLetchie, O.R., Heffernan, L.P.M., Hayn, O.A., Ellenberger, H.A., McCurdy, R.F., & Thiebaux, H.J. (1977) Electromyographic abnormalities in chronic environmental arsenicalism. *Journal of Analytical Toxicology 1,* 270–276.

Klasing, S. A., & Brodberg, R. (2003) Evaluation of Potential Health Effects of Eating Fish from Selected Water Bodies in the Northern Sierra Nevada Foothills (Nevada, Placer, and Yuba Counties): Guidelines for Fish Consumption. Pesticide and Environmental Toxicology Section, Office of Environmental Health Hazard Assessment, California Environmental Protection Agency.

Klasing, S., & Brodberg, R. (2004) Fish consumption guidelines for Lake Natoma (including nearby creeks and ponds) and the lower American River (Sacramento County): California Office of Environmental Health Hazard Assessment, 41 p.

Koch, I., Dee, J., House, K., Sui, J., Zhang, J., McKnight-Whitford, A., Reimer, K.J. (2013) Bioaccessibility and speciation of arsenic in country foods from contaminated sites in Canada. *Science of the Total Environment,* 449, 1–8.

Koch, I., McPherson, K., Smith, P., Easton, L., Doe, K. G., & Reimer, K. J. (2007) Arsenic bioaccessibility and speciation in clams and seaweed from a contaminated marine environment. *Marine Pollution Bulletin*, *54*(5), 586–594.

Kontak, D.J. & Jackson, S. J. (1999) Documentation of variable trace- and rare earth element abundances in carbonates from auriferous quartz veins in Meguma lode gold deposits Nova Scotia. *The Canadian Mineralogist*, *37*, 469–48.

Lane, P.A., Crowell, M.J., & Graves, M.C. (1988) Heavy metal removal from gold mining and tailing effluents using indigeneous aquatic macrophytes (Phase 1). *In:* Proceedings of the 5th Annual General Meeting of BIOMINET. Edited by R.G.L. McCready. CANMET SP88-23, pp. 3–37.

Lane, P.A., Pett, R.J., Crowell, M.J., MacKinnon, D.S., Graves, M.C. (1989) Further studies on the distribution and fate of arsenic and mercury at a site contaminated by abandoned gold mine tailings. *In:* Proceedings of the 6th Annual General Meeting of BIOMINET. pp. 78–107.

Lavoie, R. A., Jardine, T. D., Chumchal, M. M., Kidd, K. A., & Campbell, L. M. (2013) Biomagnification of Mercury in Aquatic Food Webs: A Worldwide Meta-Analysis. *Environmental Science & Technology*, *47*(23), 13385–13394.

LeBlanc, J.E. and Halfyard. E. A (2010) Mercury and Arsenic Concentrations in Fish Tissues and the Influence of Historic Gold Mines in Nova Scotia. Inland Fisheries Division, Nova Scotia Department of Fisheries and Aquaculture. Pictou, Nova Scotia.

Lengke, M.F., Sanpawanitchakit, C., and Tempel, R.N. (2009) The oxidation and dissolution of arsenic-bearing sulfides. *The Canadian Mineralogist*, 47, 593-613.

Little, M. E., Burgess, N. M., Broders, H. G., & Campbell, L. M. (2015) Distribution of mercury in archived fur from little brown bats across Atlantic Canada. *Environmental Pollution*, 207, 52–58.

Malcolm, W. (1929) Gold Fields of Nova Scotia. Geological Survey of Canada, Memoir 156 (reprinted in 1976 as Memoir 385), 253 p.

Mass, M. J., Tennant, A., Roop, B. C., Cullen, W. R., Styblo, M., Thomas, D. J., & Kligerman, A. D. (2001) Methylated trivalent arsenic species are genotoxic. *Chemical research in toxicology*, *14*(4), 355-361.

Milligan, T.G. and Law, B.A. (2013) Contaminants at the sediment–water interface: implications for environmental impact assessment and effects monitoring. *Environmental Science and Technology*, 47 (11), 5828–5834.

Mogren, C. L., Walton, W. E., Parker, D. R., & Trumble, J. T. (2013) Trophic Transfer of Arsenic from an Aquatic Insect to Terrestrial Insect Predators. *PLOS ONE*, *8*(6), e67817.

Moriarty, M. M., Koch, I., Gordon, R. A., & Reimer, K. J. (2009) Arsenic Speciation of Terrestrial Invertebrates. *Environmental Science & Technology*, *43*(13), 4818–4823.

Moriarty, M. M., Koch, I., & Reimer, K. J. (2011) Arsenic Speciation, Distribution, and Bioaccessibility in Shrews and Their Food. *Archives of Environmental Contamination and Toxicology*, *62*(3), 529–538.

Moriarty, M. M., Koch, I., & Reimer, K. J. (2013) Arsenic species and uptake in amphibians (*Rana clamitans* and *Bufo americanus*), *15*(8), 1520–1528.

Ng, J. C. (2005). Environmental Contamination of Arsenic and its Toxicological Impact on Humans. *Environmental Chemistry*, *2*(3), 146–160.

Nova Scotia Department of Fisheries and Aquaculture (2018) Nova Scotia Anglers' Handbook & Summary of Regulations. Available online at https://novascotia.ca/fish/documents/Anglers-Handbook-2017.pdf

Nova Scotia Department of Natural Resources (NSDNR) (2013) Species at Risk Overview. Retrieved from https://novascotia.ca/natr/wildlife/biodiversity/species-list.asp (June 2 2018).

Nova Scotia Environment (2017) Contaminated Sites: Historic Gold Mine Tailings. Available online at: https://novascotia.ca/nse/contaminatedsites/goldmines.asp.

Office of Environmental Health Hazard Assessment (OEHHA) (2018) Fish Advisories. Retrieved from: https://oehha.ca.gov/fish/advisories (April 10 2018)

O'Driscoll, N. J., Rencz, A. N., & Lean, D. R. (2005) *Mercury cycling in a wetland-dominated ecosystem: a multidisciplinary study*. Society of Environmental Toxicology and Chemistry (SETAC), 392 p.

Oremland, R.S. & Stolz, J.F. (2003) The ecology of arsenic. *Science 300*, 393-944.

Parsons, M. B., LeBlanc, K. W. G., Hall, G. E. M., Sangster, A. L., Vaive, J. E., & Pelchat, P. (2012) Environmental geochemistry of tailings, sediments and surface waters collected from 14 historical gold mining districts in Nova Scotia. *Geological Survey of Canada, Open File 7150*.

Parsons, M.B., Milligan, T.G., Yeats, P.A., Smith, J.N., Little, M.E., Parrott, D.R. (2008) Marine environmental impacts of historical gold mining activities, Isaacs, Seal, and Wine harbours, Nova Scotia. ACCESS Conference 2008. Atlantic Canada Coastal and Estuarine Science Society, Dartmouth, NS, 21–22.

Parsons, M.B. & Percival, J.B. (2005) A brief history of mercury and its environmental impact. *In* Mercury: Sources, Measurements, Cycles, and Effects; (eds.) M.B. Parsons and J.B. Percival, *Mineralogical Association of Canada Short Course, 34*, 1-20.

Park, J.-D., & Zheng, W. (2012) Human Exposure and Health Effects of Inorganic and Elemental Mercury. *Journal of Preventive Medicine and Public Health, 45*(6), 344–352.

Rahman, M.A., & Hasegawa, H. (2012) Arsenic in freshwater systems: Influence of eutrophication on occurrence, distribution, speciation, and bioaccumulation. *Applied Geochemistry, 27*(1), 304–314.

Robinson, J. (2016) Mercury concentrations in invertebrates from a contaminated wetland, Montague Gold Mines, Dartmouth, Nova Scotia. Unpublished B.Sc. Honours Thesis, Saint Mary’s University, Halifax, NS.

Saunders, J. R., Hough, C., Knopper, L. D., Koch, I., & Reimer, K. J. (2011) Arsenic transformations in terrestrial small mammal food chains from contaminated sites in Canada. *Journal of Environmental Monitoring*, *13*(6), 1784–1792.

Saunders, J. R., Knopper, L. D., Koch, I., & Reimer, K. J. (2010) Arsenic transformations and biomarkers in meadow voles (*Microtus pennsylvanicus*) living on an abandoned gold mine site in Montague, Nova Scotia, Canada. *The Science of the Total Environment*, *408*(4), 829–835.

Slotton, D. G., Reuter, J. E., & Goldman, C. R. (1995) Mercury uptake patterns of biota in a seasonally anoxic northern California reservoir. *Water, Air, and Soil Pollution*, *80*(1-4), 841-850.

Spencer, S. H., Shutler, D., & O'Brien, M. S. (2011) Correlates of mercury in female river otters (*Lontra canadensis*) from Nova Scotia, Canada. *Environmental Toxicology and Chemistry*, *30*(8), 1879–1884.

Tetford, G. (1999) The problem of mercury in the environment: abandoned gold mines in Nova Scotia. Unpublished B.Sc. Honours Thesis, Saint Mary's University, Halifax, NS.

Tweedy, B. N., Drenner, R. W., Chumchal, M. M., & Kennedy, J. H. (2013). Effects of Fish on Emergent Insect-Mediated Flux of Methyl Mercury across a Gradient of Contamination. *Environmental Science & Technology*, 47(3), 1614–1619.

Walker, T.R. & Grant, J. (2015) Metal(loid)s in sediment, lobster and mussel tissues near historical gold mine sites. *Marine Pollution Bulletin*, 101, 404–408.

Whaley-Martin, K. J., Koch, I., Moriarty, M., & Reimer, K. J. (2012) Arsenic Speciation in Blue Mussels (*Mytilus edulis*) Along a Highly Contaminated Arsenic Gradient. *Environmental Science & Technology*, *46*(6), 3110–3118.

Whaley-Martin, K. J., Koch, I., & Reimer, K. J. (2013) Determination of arsenic species in edible periwinkles (*Littorina littorea*) by HPLC–ICPMS and XAS along a contamination gradient. *Science of The Total Environment*, 456–457 (Supplement C), 148–153.

Wong, H. K. T., Gauthier, A., & Nriagu, J. O. (1999) Dispersion and toxicity of metals from abandoned gold mine tailings at Goldenville, Nova Scotia, Canada. *Science of The Total Environment*, *228*(1), 35–47.

Table 1. Timeline and summary of the 23 articles included in this literature review describing [Hg] and/or [As] in ecological receptors at (or in association with) historical gold mine districts in Nova Scotia.

Article	Gold District(s)	Taxonomic Group(s)	Sample Information
Eaton (1978)	Montague, Mount Uniacke, Oldham	Aquatic Plants (*n*=40) Terrestrial Plants (*n*=53) Freshwater Invertebrates (*n*=3) Amphibians (*n*=17) Fish (*n*=28) Mammals (mice) (*n*=4)	• Identified to common names • Sample tissue not specified
Brooks et al. (1981)	Montague, Moose River, Caribou, Salmon River, Goldenville, Molega	Aquatic Plants (*n*=6) Terrestrial Plants (*n*=1)	• Identified to species • Sample tissue not specified
Brooks et al. (1982)	Montague	Freshwater Invertebrates (*n*=7, composite samples of 10) Fish (*n*=3, composite samples of 5)	• Identified to common names • Analyzed whole
Dale and Freedman (1982)	Montague	Terrestrial Plants (*n*=15, *n*=7 Ref, composite samples of 5) Fish (*n*=2)	• Identified to species • Sample tissue not specified
Eaton and Clair (1985)	Waverley	Fish (*n*=27)	• Identified to species • Analyzed filets
Lane et al. (1988)	Oldham	Freshwater plants (*n*=12, *n*=2 Ref, composite samples of 5+)	• Identified to species • Analyzed roots and shoots separately
Lane et al. (1989)	Oldham	Freshwater Plants (*n*=9, composite samples ranging from 5 to 74) Terrestrial Plants (*n*=23, composite samples ranging from 6 to 43)	• Identified to species • Sample tissue not specified
Wong et al. (1999)	Goldenville	Freshwater plants (*n*=unspecified), Terrestrial plants (*n*=unspecified)	• Identified to species • Analyzed stems and leaves • No reference data provided

Tetford (1999) Unpublished	Caribou	Fish (*n*=9, *n*=12 Ref)	• Identified to species • Sample tissue not specified
Koch et al. (2007)	Lower Seal Harbour	Marine mollusks (*n*=unspecified) Marine plants (*n*=unspecified)	• Identified to species • Sample tissue not specified • No reference data provided
Moriarty et al. (2009)	Montague, Lower Seal Harbour, Upper Seal Harbour	Terrestrial invertebrates (*n*=21, *n*=7 Ref, composite samples of unknown amount)	• Identified to order • Analyzed whole
Saunders et al. (2010)	Montague	Mammals (voles) (*n*=10, *n*=10 Ref)	• Identified to species • Analyzed by tissue type (stomach components, digestive, non-digestive, liver)
LeBlanc and Halfyard (2010)	Montague, Lower Seal Harbour, Moose River, Molega, Lake Charlotte, Waverley, Caribou	Fish (*n*=184, *n*=51 Ref)	• Identified to species • Sample tissue not specified
Saunders et al. (2011)	Upper Seal Harbour, Lower Seal Harbour	Terrestrial plants (*n*=12, *n*=9 Ref) Mammals (voles) (*n*=17, *n*=22 Ref)	• Identified to species • Analyzed by tissue type (stomach components, digestive, non-digestive, liver)
Moriarty et al. (2011)	Upper Seal Harbour	Mammals (shrews) (*n*=12, *n*=9 Ref)	• Identified to species • Analyzed whole
Button et al. (2012)	Lower Seal Harbour	Terrestrial invertebrates (*n*=unspecified) Leaf Litter (*n*=unspecified)	• Invertebrates identified to species • Analyzed whole
Whaley-Martin et al. (2012)	Lower Seal Harbour	Marine Mollusks (*n*=4, composite samples of 10-15, *n*=2 Ref, composite samples of 13-15)	• Identified to species • Analyzed whole (shells removed)
Koch et al. (2013)	Lower Seal Harbour	Terrestrial plants (berries) (*n*=4, *n*=2 Ref) Fungi (*n*=2) Mammals (hares) (*n*=2, *n*=1 Ref)	• Identified to common names, except mushrooms to species • Sample tissue not specified
Whaley-Martin et al. (2013)	Lower Seal Harbour	Marine Mollusks (*n*=4, composite samples of 100-201, *n*=1 Ref, composite samples of 100-203)	• Identified to species • Analyzed whole (shells removed)
Moriarty et al. (2013)	Upper Seal Harbour	Amphibians (*n*=7, *n*=5 Ref)	• Identified to species • Analyzed legs

Walker and Grant (2015)	Lower Seal Harbour, Isaacs Harbour	Marine Shellfish (*n*=unspecified, *n*=40 Ref)	• Identified to species • Analyzed lobster hepatopancreas, mussles whole (shell removed)
Robinson (2016) Unpublished	Montague	Terrestrial Invertebrates (*n*=73+, *n*=47 Ref)	• Identified to genus or suborder • Analyzed whole
Doe et al. (2017)	Lower Seal Harbour, Goldenville, Gold River, Harrigans Cove, Lawrencetown, Salmon River, Tangier	Marine Mollusks (*n*=20, *n*=5 Ref)	• Identified to species • Analyzed mollusk tissue

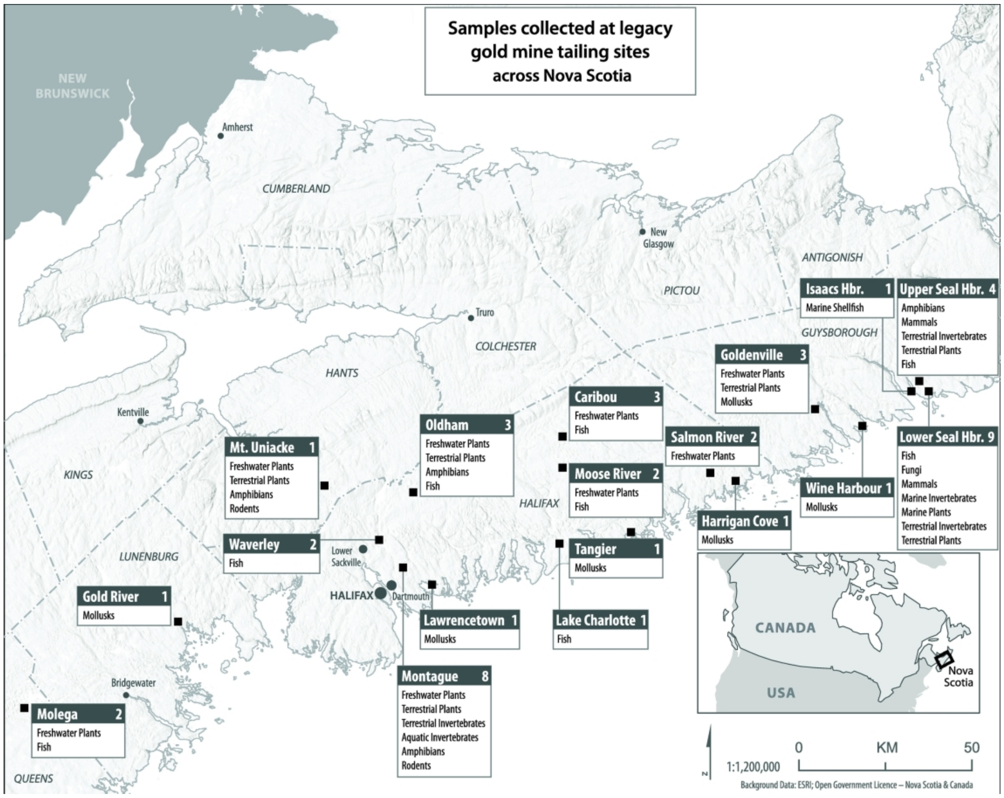

Figure 1. Summary map of historical gold mine districts, and associated broad taxonomic groups, included in literature review of tailings-related Hg and As accumulation. See Table 1 for data breakdown.

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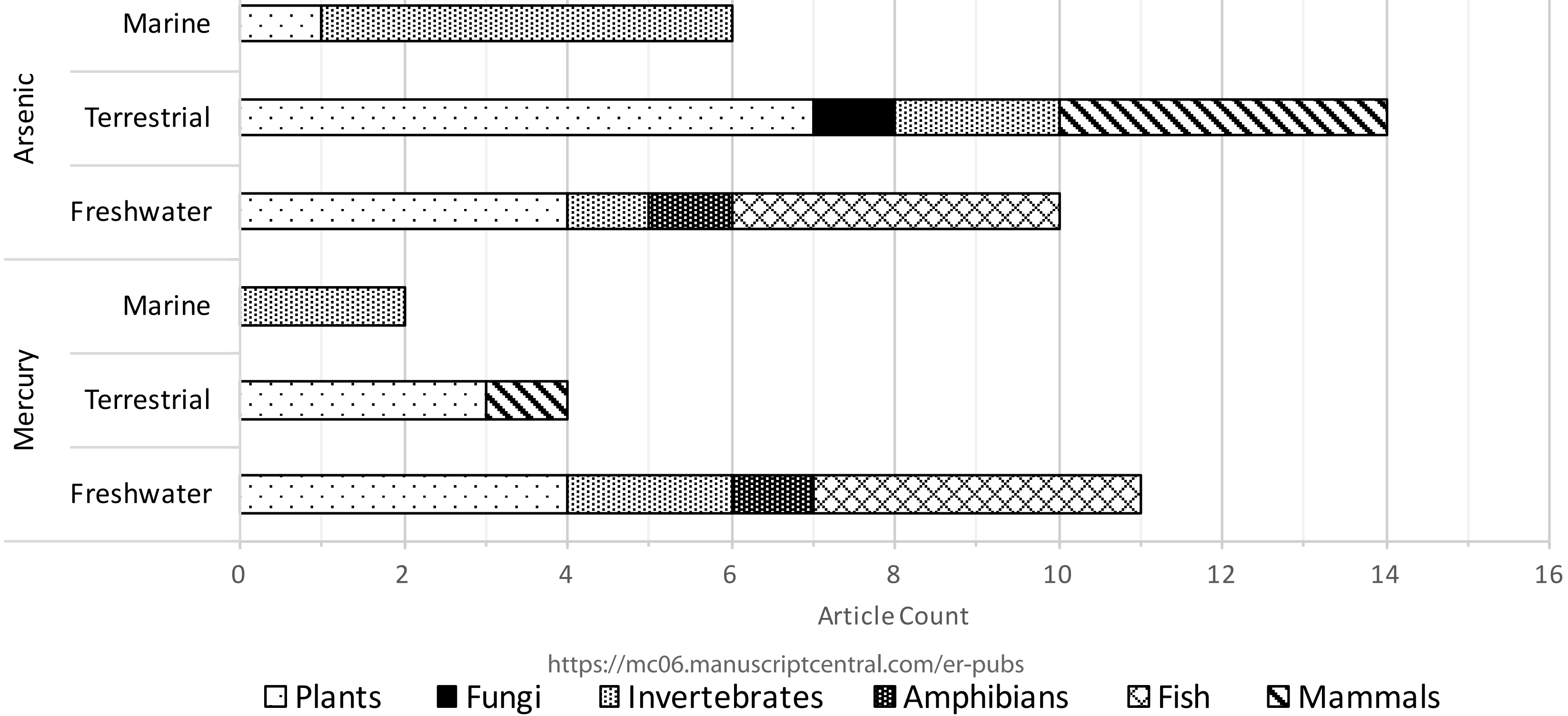
Arsenic
Marine
Terrestrial
Freshwater
Mercury
Marine
Terrestrial
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0
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Invertebrates
Amphibians
Fish
Mammals

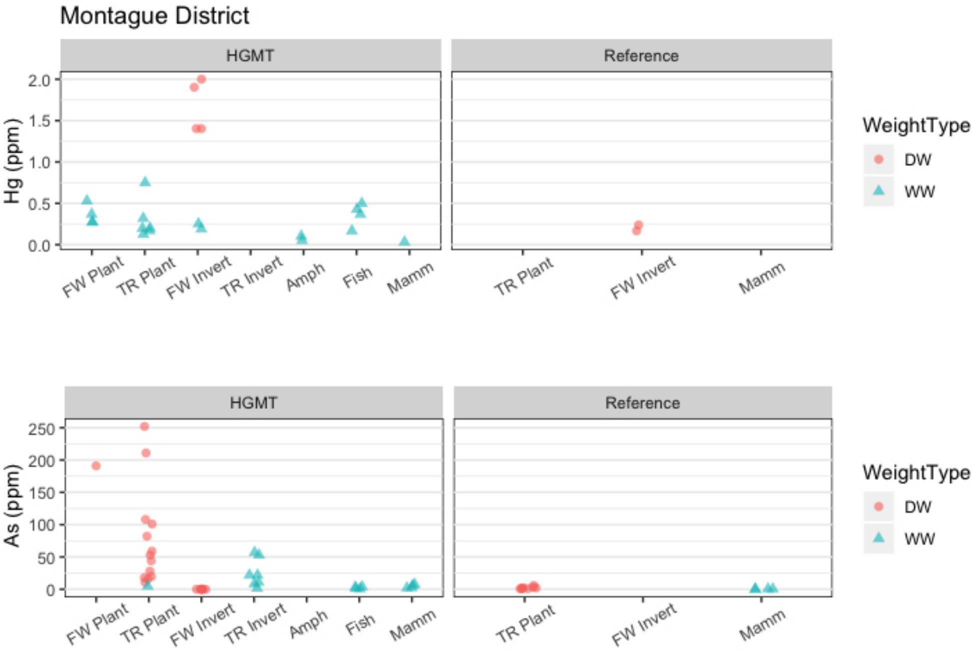

Figure 3. Summary of [Hg] and [As] data collected at the Lower Seal Harbour historical gold mining district, the most studied site within Nova Scotia. Data points are separated by wet weight (ww) and dry weight (dw). Sources are described in Table 1. HGMT = historical gold mine tailings; TR = terrestrial; MA = marine.

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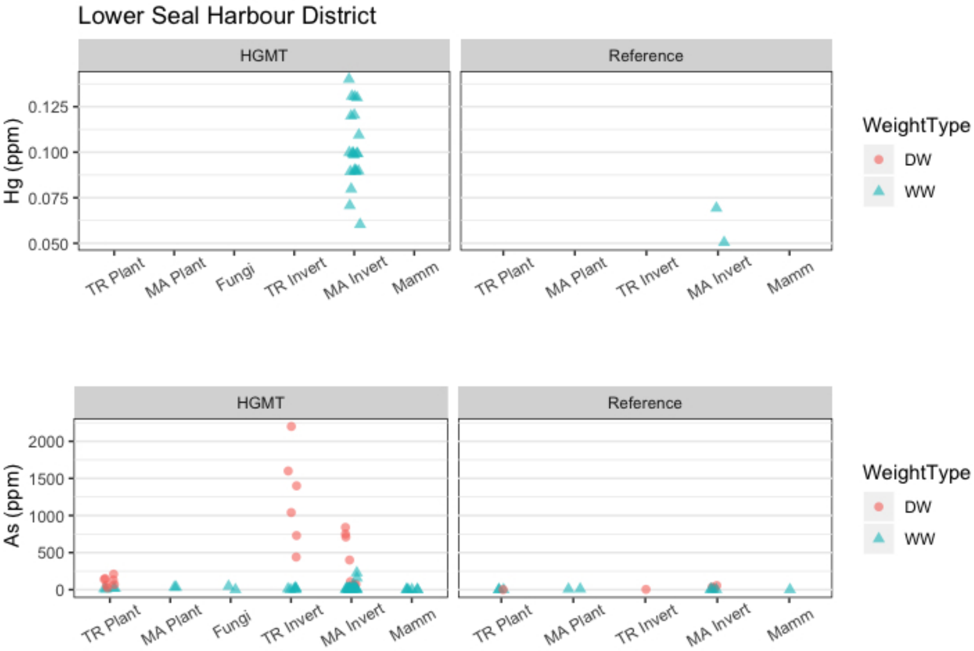

Figure 4. Summary of [Hg] and [As] data collected at the Montague historical gold mining district, the second most studied site within Nova Scotia. Data points are separated by wet weight (ww) and dry weight (dw). Sources are described in Table 1. One outlier As concentration from Dale and Freedman (1982) removed (slender rush, 834 ppm As). HGMT = historical gold mine tailings; TR = terrestrial; MA = marine.

254x176mm (72 x 72 DPI)