ARCTIC
VOL. 76, NO. 4 (DECEMBER 2023)

InfoNorth

Seasons of Change: Ecological Recovery From Legacy Arsenic Pollution in Rapidly Warming Subarctic Lakes

by Amanda Little

INTRODUCTION

Located 5 km north of the City of Yellowknife (Northwest Territories), the abandoned Giant Mine has received an updated remediation bill estimated at $4.38 billion (Cohen, 2022), making it Canada's most expensive remediation project to date (Aboriginal Affairs and Northern Development Canada, 2012; Blake, 2022). Over 237,000 tonnes of toxic arsenic trioxide dust are approximated to be buried underneath the degrading mine, in addition to the ~20,000 tonnes of arsenic that was released into the surrounding area between 1948 and 1999 (Wrye, 2008; Houben et al., 2016). Lakes within a 30 km radius have been heavily impacted (Hocking et al., 1978; Hutchinson et al., 1982; Galloway et al., 2015; Walker et al., 2015; Palmer et al., 2019; Jasiak et al., 2021), with some lakes exhibiting arsenic concentrations 400 times greater than the 5 µg/L guideline for the protection of aquatic life (Wagemann et al., 1978; CCME, 2001; Sivarajah et al., 2019). Preliminary studies show that minimal biological recovery has occurred in many of the lakes in the region despite modern day arsenic concentrations being lower than they were during periods of mining (Thienpont et al., 2016). Yellowknife is also experiencing the combined effects of accelerated climate change and urbanization, which complicates recovery from legacy arsenic pollution (Cai, 2005; Stewart et al., 2018). Changing temperature and precipitation regimes are expected to result in dramatic changes in the region, impacting critical biogeochemical cycles, including arsenic (MacDonald et al., 2005; Astles et al., 2022). There is currently no remediation plan for the region outside of the property's perimeter, and therefore an improved understanding of the processes that underlay lake ecological recovery will be critical as communities navigate decisions about the future use of these areas.

The contamination of water bodies by arsenic is an issue of increasing global concern due to the chemical's widespread distribution and overall toxicity (Wagemann et al., 1978; Wang and Mulligan, 2006). In aquatic systems, arsenic is typically found as one of two inorganic chemical species—arsenate [As(V)] and arsenite [As(III)] (Fig. 1). In general, inorganic arsenic species are considered to be more toxic than their organic counterparts, with some studies reporting decreases in toxicity ranging between 100–1000 times for organic species (i.e., dimethylarsinic acid [DMA(V)] and trimethylarsenic oxide [TMAO(V)]) relative to inorganic As(III) (Hirano et al., 2004). The generally accepted relative toxicity of arsenic species to freshwater organisms from most to least toxic is: dimethylarsinous acid [DMA(III)] = monomethylarsonous acid [MMA(III)] > As(III) > As(V) > monomethylarsonic acid [MMA(V)] = DMA(V) > TMAO(V), with trivalent species considered to be more toxic than their pentavalent counterparts (Akter et al., 2005). Despite the differences in toxicity between arsenic species, both the current drinking water and protection of aquatic life guidelines have been set at 10 µg/L and 5 µg/L, respectively, for total freshwater arsenic concentrations (CCME, 2001; WHO, 2017).

The chemical form in which arsenic is found is largely determined by the environmental conditions present in aquatic systems. Arsenic is a redox-sensitive element, which means that it will undergo chemical transformations depending on the amount of dissolved oxygen (DO) available, and therefore the reducing or oxidizing conditions present. Under oxidizing conditions, As(V) is the dominant arsenic ion, while reducing conditions host higher proportions of As(III) (Cullen and Reimer, 1989). As(V) tends to be the most common form of arsenic found in freshwater systems. The redox conditions of aquatic systems are dependent on the amount of DO present, the position of the redoxcline in relation to the sediment-water interface (SWI), the availability of electron acceptors (e.g., O_2, NO_3^-, SO_4^{2-}) and donors (e.g., NH_4^+, H_2S, CH_4, Fe^{2+}), and the rates of microbial activity and organic matter deposition (Rodie et al., 1995). Under reducing conditions, and particularly in the presence of an electron donor like organic matter, the reduction of As(V) to As(III) is thermodynamically favourable, with the opposite reaction favoured under oxidizing conditions (Rodie et al., 1995). Many of these redox-based chemical transformations occur in either near-surface zones (i.e., surface waters) or at the SWI (i.e., bottom waters). These zones, and their subsequent processes, are critical controls of arsenic mobility and concentration, as the metalloid can both be captured from and released back into the water column depending on the environmental conditions present. Therefore, the chemical

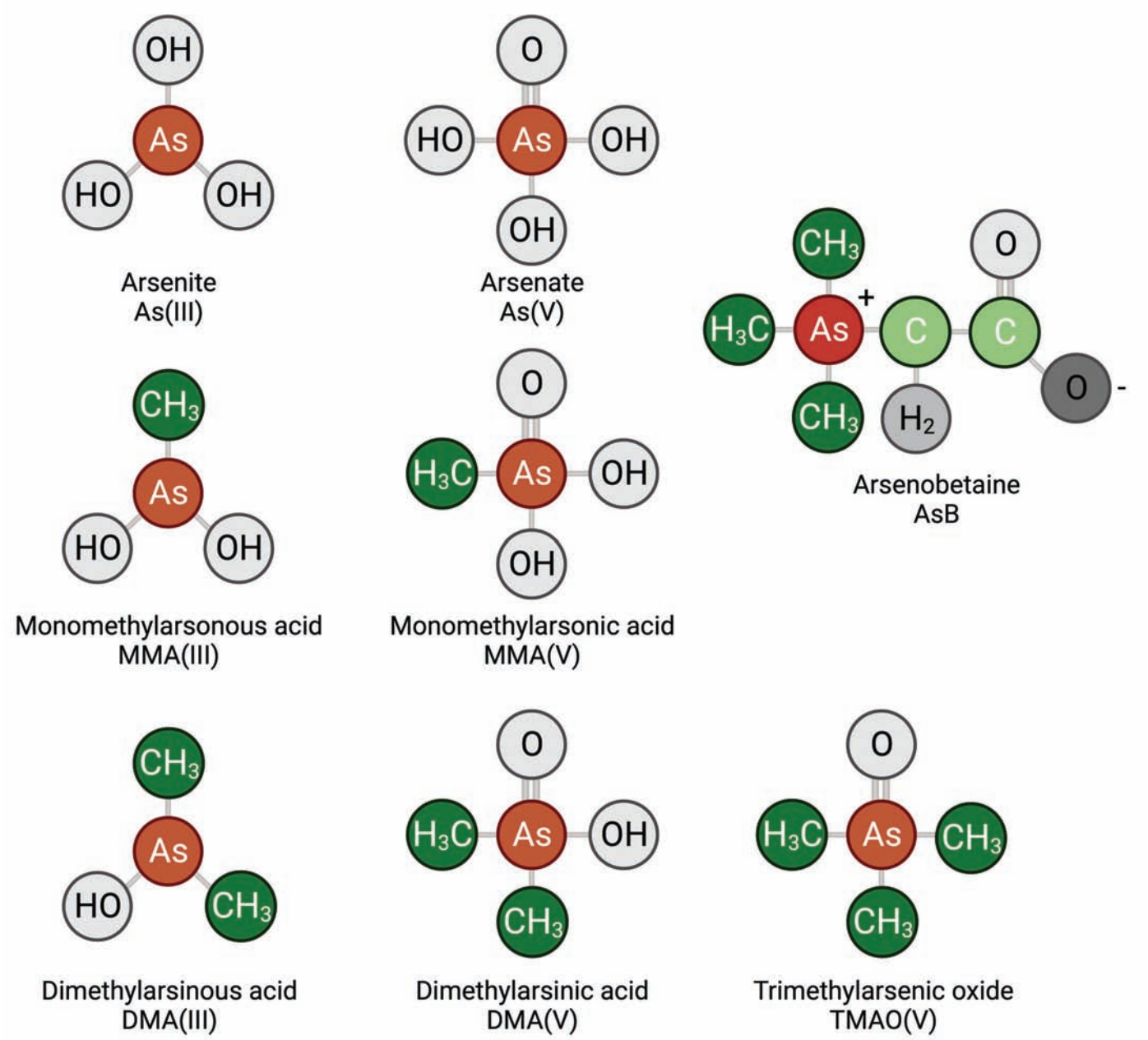

FIG. 1. Chemical structures of common arsenic species in aquatic systems. Created with BioRender.com.

structure of arsenic present in aquatic ecosystems can have profound impacts on the contaminant's overall mobility, concentration, bioavailability, and subsequent toxicity, with important implications for timelines of recovery from legacy arsenic pollution.

In addition to DO, arsenic speciation, mobility, and toxicity have been shown to be influenced by dissolved organic matter (DOM) and phosphorus. DOM and phosphorus are of particular interest since both have been observed to be impacted by watershed development and climate change in the Yellowknife region (Pienitz et al., 1999; Stewart et al., 2018; Sivarajah et al., 2020, 2021). DOM impacts arsenic by both competing for, but also providing, active sites for arsenic to bind with, depending on the type of organic matter and environmental conditions (Zhang et al., 2014; Pothier et al., 2020). The presence of DOM can therefore result in either increases or decreases of arsenic concentration in the water column, particularly near the SWI or other regions where DO may be depleted. Similarly, phosphorus impacts arsenic due to the fact that its most common environmental form, phosphate (PO_4^{3-}), is a chemical analogue of As(V) and can therefore compete with it for sorption sites (Cullen et al., 1994; Levy et al., 2005; Zhang et al., 2014; Wang et al., 2015). This works both ways, however, as As(V) can use the same channels as PO_4^{3-} to enter the cells of exposed organisms, therefore increasing arsenic's toxicity to certain life forms (Hellweger et al., 2003; Levy et al., 2005). As a result, systems experiencing

changes in DOM and PO_4^{3-} concentrations exhibit complex relationships with arsenic biogeochemical cycling. However, it should be noted that higher concentrations of arsenic in the water column as a result of DOM or PO_4^{3-} influence does not always result in arsenic becoming more bioavailable, as additional competition for uptake by phytoplankton exists.

Finally, depth is also an important control of arsenic cycling as it determines whether a lake will thermally stratify (the formation of distinct layers of water based on temperature and density) or experience episodic stratification events during the summer and winter months. Deep lakes are more likely to undergo thermal stratification, which results in the bottom layers of water being cut off from incoming sources of oxygen from the atmosphere. As a result, DO is consumed over time, leading to reducing conditions that cause arsenic to be released from the sediment back into the water column as As(III) (Rodie et al., 1995); However, because of the density difference between the top and bottom of the lake (i.e., epilimnion and hypolimnion), this As(III) cannot make its way to the top of the lake, and it instead builds up over time in the hypolimnion. This situation can lead to significant concentration and toxicity gradients with depth, as more arsenic is released and then trapped at the bottom of the lake in the more toxic As(III) form, compared to the surface of the lake where the majority of arsenic is likely to be found as As(V) due to the higher DO content. Therefore, plankton and other aquatic organisms that are typically found in surface waters or cannot swim in and out of the hypolimnion tend to have lower levels of arsenic in their bodies (Vaquer-Sunyer and Duarte, 2008; Barrett et al., 2018). However, the release of arsenic from lake sediments is a kinetically-mediated process and therefore temperature dependent (Ratkowsky et al., 1982; Weber et al., 2010; Barrett et al., 2019). This means that during periods of low DO availability, shallow, well-mixed lakes may actually host higher concentrations of arsenic than the hypolimnions of deep lakes because of the warming of the top layer of sediments by the sun. Subsequently, higher concentrations and proportions of total arsenic have been observed in the water and plankton communities of shallow, well-mixed lakes (Barrett et al., 2018).

When all three of these environmental variables are considered in tandem, it becomes clear that climate change has substantial implications for recovery from legacy arsenic pollution in Yellowknife lakes, and that arsenic concentrations can vary drastically depending on the time of year. This is a critical issue in limnology, as lakes are typically only sampled during the open water season. For example, since arsenic biogeochemical cycling is intrinsically linked to DO availability, environmental controllers of DO also influence arsenic. In fact, a 2019 study by Palmer et al. observed a 4-fold increase in arsenic concentration (172 – 846 µg/L) in a shallow lake from the Yellowknife region, with the highest levels occurring in tandem with DO depletion. Important seasonal phenomena which affect DO include ice cover, hydrological connectivity, and inputs of nutrients and organic matter. All these seasonal features are predicted to shift under climate change, and as a result, so will DO availability and subsequently arsenic concentration and toxicity. In particular, since arsenic tends to be released back into the water column as the more toxic As(III) under low DO (anoxia) and reducing conditions, it can be reasoned that certain periods of the year may pose a higher toxicological risk for exposure to arsenic in lakes (such as late summer and winter), and that this risk may increase under climate change with warming temperatures and prolonged periods of anoxia.

While arsenic concentration and speciation appear to be strongly influenced by seasonal factors, plankton community structure and size are as well. The most important and universally recognized seasonal controls on plankton succession are temperature, light attenuation, and nutrient concentrations (Sommer et al., 1986, 2012). Plankton and other aquatic biological organisms have also been shown to be important detoxification pathways for different arsenic species (Caumette et al., 2011, 2014; Rahman et al., 2014). One such pathway—the biomethylation transformation of inorganic to organic arsenic species—is mediated by plankton and other microorganisms and appears to be particularly common in phytoplankton (Navratilova et al., 2011; Rahman and Hasegawa, 2012; Price et al., 2012). In addition, a study by Caumette et al. (2011) found that phytoplankton had significantly higher proportions of arsenic compared to zooplankton. However, they also observed that lakes with lower arsenic concentrations tended to host larger zooplankton individuals compared to those with higher concentrations and had a higher proportion of organic arsenic species (Caumette et al., 2011). Therefore, while seasonal factors such as temperature and DO availability control the concentration and speciation of arsenic in the water column, the plankton community present in a lake may also be an important influence and will likely also be influenced by the combined stressors of watershed development and climate change in the Yellowknife region (Fig. 2).

To be able to successfully predict how arsenic and plankton communities will change in the future, it's important to have high resolution, long-term data to assess how they have behaved in the past. Unfortunately, long-term environmental datasets are hard to come by, which is why paleolimnological techniques have become so widespread in limnological research. Paleolimnology is the reconstruction of past environmental conditions through the analysis of a combination of biological, chemical, and physical proxies as captured and stored in lake sediments. When assessing the long-term impacts of arsenic contamination in the Yellowknife region, it's reasonable to assume that lakes located near the Giant Mine site received higher amounts of arsenic and other mining-related contaminants compared to those farther away. Although

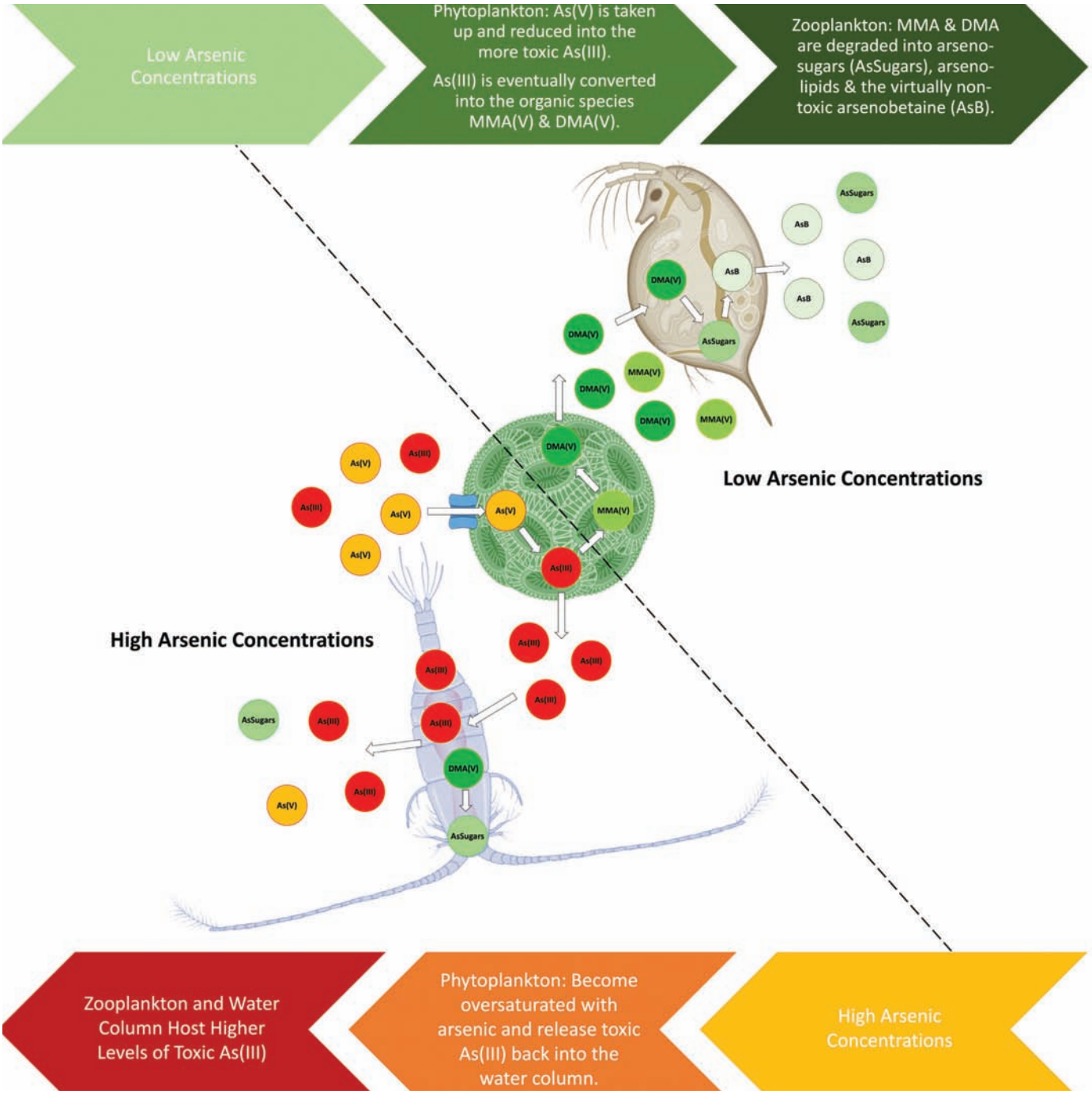

FIG. 2. Proposed detoxification pathway of arsenic in aquatic systems via biomethylation in phytoplankton. Arsenic enters the cell through phosphate transporters as As(V) and is rapidly transformed into As(III). Under low arsenic concentrations, As(III) is then converted into a significantly less toxic, organic form of arsenic before being excreted back into the water column. Under high arsenic concentrations, As(V) enters the cell the same way and is rapidly transformed into As(III). However, under these conditions, the cell quickly becomes oversaturated with arsenic and the toxic As(III) is forced back into the water column due to concentration gradients between the cell and surrounding environment. Created with BioRender.com.

the toxicity endpoints calculated from observations made under laboratory conditions tend be much higher than concentrations typically found in real world settings, a few paleolimnological investigations have found that zooplankton communities have responded to changes in arsenic levels. Coinciding with the onset of mining inputs, studies from the Yellowknife region (Thienpont et al., 2016), southwest China (Chen et al., 2015), and southern Ontario (Tenkouano et al., 2019) all indicate significant community assemblage changes, if not outright extirpation of local phyto- and zooplankton, in response to the input of arsenic and other contaminants. While it is important to note that

these communities are likely responding to several different stressors, the changes in arsenic concentrations were all significantly higher than other chemicals (e.g., 1700% increase in arsenic by weight in the sediment core collected by Thienpont et al., 2016) and, in some cases, surpassed the known lab-based toxicity endpoints for plankton. Total arsenic concentrations are therefore occurring in nature at levels known to cause ecotoxicological harm. This is significant because phytoplankton make up the base of the aquatic food web while zooplankton play an intermediary role in ecosystem structure, acting as grazers and predators for the trophic level immediately below them and as prey for the level immediately above. Therefore, a significant change in the overall biomass of plankton is likely to lead to direct and indirect effects on the rest of the ecosystem.

Bringing all of these ideas together, my PhD research aims to address the research gaps surrounding controls of arsenic biogeochemical cycling with emphasis on aquatic ecotoxicity and ecological recovery by exploring three main themes: 1) modern plankton community structure across spatial gradients of arsenic, DOM, PO_4^{3-}, and depth; 2) seasonal aquatic cycling of arsenic and the subsequent impacts on plankton communities; and 3) how arsenic exposure has shaped lake community structures through time. This research will combine methods from paleolimnology, biogeochemistry, ecology, and ecotoxicology to investigate how arsenic biogeochemical cycling, mobility, and toxicity shift in response to changing seasonal and climatic influences, what the potential implications are for both human and aquatic ecosystem health, and how to tackle the complex challenge of remediating legacy arsenic pollution in rapidly changing northern landscapes.

RESEARCH APPROACH

To address my research themes, I have been collecting samples on three temporal scales: 1) present-day snapshots; 2) interannual; and 3) interdecadal. Due to the frequency with which samples need to be collected, I have been working closely with Dr. Michael Palmer and colleagues from the Aurora Research Institute who are based in Yellowknife.

For our present-day snapshots, we sampled 50 lakes in the Yellowknife region via helicopter in June 2022. This was done to create a robust spatial dataset to represent how arsenic continues to impact modern day lake ecosystems. Our guiding objectives were to: 1) characterize summer phytoplankton and zooplankton community assemblages in order to determine the regional biogeography of these groups; 2) determine if plankton communities with higher arsenic concentrations have lower overall abundance and diversity than systems with lower concentrations; and 3) investigate the role of DOM, PO_4^{3-}, and lake depth in modulating the relationship (or lack thereof) between arsenic and plankton community structure.

The 50 lakes were selected based on previous sampling efforts completed in 2012 and 2014 (Palmer et al., 2016), allowing me to compare arsenic concentrations nearly a decade later and to investigate the impacts of additional controlling variables. We established water column profiles using a multi-parameter probe, with emphasis on conductivity, temperature, pH, and DO. Water samples were collected for a general suite of water quality parameters, including total arsenic, phosphorus, and chlorophyll *a*. Separate samples were collected for water arsenic speciation data via a vertical Van Dorn from the top 1 m of each lake. Phytoplankton were collected using this same Van Dorn, and zooplankton were collected via vertical net tows using a Wisconsin net. Surface sediment grabs were collected using an Ekman Grab from lakes with depths between 3 and 10 meters to determine fish presence/absence based on the subfossil remains of *Chaoborus* spp. (phantom midges), as some species of Chaoborus are unable to coexist with fish (Sweetman and Smol, 2006). Lakes shallower than 3 meters are assumed to be fishless, while lakes deeper than 10 meters are assumed to contain fish. This will help us to disentangle the potential effects of grazing from fish versus arsenic exposure on zooplankton communities in the region. This data will be used to make predictive models (e.g., Vucic et al., 2020) about how arsenic impacts plankton communities, and by extension, entire aquatic ecosystem health.

The interannual perspective focuses on the seasonal changes in arsenic biogeochemical cycling in Yellowknife lakes. I'm interested in determining how arsenic concentration and speciation change depending on the time of year, and the subsequent interactions with phytoplankton and zooplankton biomass, diversity, and abundance. I'm particularly interested to see whether deeper lakes with lower arsenic concentrations host plankton communities with lower overall arsenic body burden, but with higher proportions of organic arsenic species, specifically arsenosugars, arsenolipids, and arsenobetaine. I'm also interested in investigating how both arsenic and plankton communities behave during the shoulder seasons (i.e., spring, fall), particularly during spring thaw, as these periods are understudied because of the difficulties associated with working safely on lake ice. I expect to find higher arsenic concentrations and proportions of As(III) near the SWI in the shoulder seasons due to DO consumption over the period of prolonged ice cover (late winter/early spring) and organic matter decomposition from enhanced summer lake productivity (fall).

To explore these ideas, we selected five study lakes in the Yellowknife region. All five were sampled in April, May, June, September, and November of 2022, and again in February, April, and May of 2023. Water sampling included depth profile analysis at one-meter intervals, deployment of continuous loggers to measure temperature and DO, total arsenic and arsenic speciation, chlorophyll *a*, and DOM concentrations. Phytoplankton and zooplankton were collected for community assemblage data with

additional bulk samples collected to determine total and chemical species-specific body arsenic burden at each trophic level via ICP-MS and HPLC-ICP-MS analysis in Dr. Marc Amyot's the lab at the Université de Montréal. Special attention was given to conditions pre- and during ice breakup in April and May of 2022 and 2023. During this time, snow and ice thickness were measured, ice was assessed for signs of attached algal growth, and both water and plankton samples were collected as described above. Photosynthetically active radiation (PAR) and chlorophyll *a* were also measured throughout the water column to identify the period of rapid increase in primary productivity as under-ice light penetration increased over the late winter to early spring transition period.

My final temporal perspective is assessing plankton communities in lakes from the region prior to, during, and after exposure to mining-related arsenic contamination. I am particularly interested in determining whether lakes closer to the Giant Mine site exhibit more significant plankton community assemblage changes compared to lakes which are farther away, and to compare community assemblages between the pre-mining, during, and post-mining time periods. Building from studies by Thienpont et al. (2016) and Persaud et al. (2021), I have been investigating the impacts of arsenic, specifically on Cladocera (Branchiopoda) communities. Cladocera are a group of crustacean zooplankton that preserve well in lake sediments and can be identified to species-level under compound microscopes. I chose Cladocera as they have been relatively understudied in Yellowknife lakes, especially when compared to diatoms (Bacillariophyta). Nine lakes were selected to represent a gradient of spatial arsenic concentrations from the region, as well as depth, DOC, and phosphorus gradients. Sediment cores were collected with a gravity corer and sectioned into 1.0 cm intervals using a modified Glew (1988) extruder (Fig. 3). These cores have been dated via cold vapour atomic absorption spectrometry to estimate chemical concentrations and plankton community structure present each time frame since the beginning of mining activities and to assess baseline (i.e., pre-mining) conditions. The combination of these methods and temporal perspectives allow me to explore how arsenic has impacted aquatic ecosystems through time (interdecadal), how it changes in toxicity and mobility at different times of the year (interannual), and the current spatial gradient of arsenic contamination in the region (present-day snapshots).

FIG. 3. Amanda Little holding a sediment core taken from Frame Lake. Sediment cores can be sectioned into discrete intervals and then analyzed under the microscope for various biological, chemical, and physical properties, which allows scientists to reconstruct historical environmental conditions. This sediment core was sectioned into 1.0 cm intervals and investigated for Cladocera community and chemical changes.

SIGNIFICANCE

While arsenic is a contaminant of increasing global concern, little is known about its long-term impacts in the environment and how it is biogeochemically transformed by the aquatic organisms it interacts with. This is particularly evident when considering how little we know about the hypothesized detoxification pathway performed by phytoplankton and other lower trophic level organisms through the conversion from more toxic inorganic arsenic species into virtually non-toxic organic species. Since arsenic is a redox-sensitive element, we can predict that changes in oxygen availability in northern lakes, as the combined result of anthropogenic climate change and watershed development, will likely impact how arsenic behaves in these systems. Certain times of the year may pose a higher health risk to both human and non-human life due to increases in arsenic concentration and overall toxicity because of its chemical form. Therefore, having a better understanding of natural detoxification processes and better characterizing how arsenic cycles and impacts surrounding ecosystems across a variety of temporal scales is critical to discerning how it will behave in the future under a rapidly warming climate.

This research provides novel information about plankton communities throughout different times of the year, including winter and spring thaw—two periods which have been understudied because of difficulties associated with ice and weather safety, particularly in subarctic and Arctic regions. It is particularly relevant when assessing current toxicological guidelines and for establishing

realistic ecosystem health baselines in regions which have been impacted by arsenic contamination for decades. It will ultimately help guide cumulative effects assessments of climate change that can consider the role of legacy pollution—a newly emerging environmental management strategy. If arsenic concentrations begin to increase as a consequence of climate change, particularly during the summer and winter months when activities such as fishing are more popular, then it makes sense that the toxicological and municipal health and safety guidelines in these regions should be updated to reflect the increased seasonal risk of arsenic exposure. Finally, a portion of my research is already being applied to test a potential remediation strategy in a lake in Yellowknife's city centre. An aerator will be installed in the lake later in 2024 in an attempt to maintain under-ice oxygen over the winter (Blake, 2021). This strategy will determine whether arsenic can be locked in place in the sediment and therefore be prevented from remobilizing into overlying waters. If successful, the installation of aerators in arsenic contaminated water bodies, including drinking water sources, may become a major remediation strategy used by gold and silver mines across the world.

ACKNOWLEDGEMENTS

I am incredibly honoured and grateful to have been selected as the recipient of the Arctic Institute of North America's 2023 Jennifer Robinson Memorial Scholarship. I would also like to thank the Yellowknives Dene First Nations for allowing me to work on their traditional lands. Additional funding and logistical support for this research was provided by NSERC (Discovery Grant to Dr. J. B. Korosi); the Weston Family Foundation (Weston Family Awards in Northern Research [Doctoral Level]); Polar Continental Shelf Program; Polar Knowledge Canada (POLAR Scholarship, Northern Science Training Program); York University (OGS; Queen Elizabeth II Graduate Scholarship in Science and Technology), Faculty of Graduate Studies (Academic Excellence Fund; Research Cost Fund), and Faculty of Environmental and Urban Change (the Adrienne and Donna Pocock Memorial Award); and the Aurora Research Institute. This research was conducted under the Aurora Research Institute's Northwest Territories Research License 16784.

I have been incredibly lucky to work with Dr. Jennifer B. Korosi as my mentor and PhD supervisor. She has been incredibly supportive and encouraging of both myself and this project throughout its entirety. I'd also like to thank Dr. Michael Palmer. Mike has been a champion of this research from the very beginning and has provided countless pep talks and brainstorm sessions (not to mention funding, equipment, personnel support, the list goes on and on), without which this project wouldn't have been possible. I have also had the pleasure of working with a number of incredible collaborators, many of whom were critical to this project's realization. I'd like to thank Dr. Derek Gray, Dr. Heidi Swanson, and Dr. Marc Amyot for their guidance and support. Thank you to Dominic Bélanger and Maria Alaoui for their work on arsenic speciation analyses and to Dr. Dominic Ponton for his advice on trace metal sampling design. Finally, I'd like to thank all of the fieldwork team members and undergraduate students who have assisted me with this research: Angela Hamilton, Nigel Rossouw, Seamus Daly, Charlie West, Sorin-Alexandru Gruia, Rachael Abulu, Rebecca Gasman, Randelle Cheyanna Adano, Victoria Carroll, Nicole Andreola, Madeline Patenall, Asli Yusuf, and Altrisha Rodrigues. Without them, this work would not have been possible.

REFERENCES

Aboriginal Affairs and Northern Development Canada. 2012. Internal audit report: Value for money audit of the Giant Mine remediation project. Project No. 12-32. Ottawa, Ontario. https://publications.gc.ca/collections/collection_2016/aanc-inac/R5-96-2012-eng.pdf

Akter, K.F., Owens, G., Davey, D.E., and Naidu, R. 2005. Arsenic speciation and toxicity in biological systems. Reviews of Environmental Contamination and Toxicology 184:97–149. https://doi.org/10.1007/0-387-27565-7_3

Astles, B.C., Chételat, J., Palmer, M.J., and Vermaire, J.C. 2022. Experimental investigation of short-term warming on arsenic flux from contaminated sediments of two well-oxygenated subarctic lakes. PLoS ONE 17(12): e0279412. https://doi.org/10.1371/journal.pone.0279412

Barrett, P.M., Hull, E.A., King, C.E., Burkart, K., Ott, K.A., Ryan, J.N., Gawel, J.E., and Neumann, R.B. 2018. Increased exposure of plankton to arsenic in contaminated weakly-stratified lakes. Science of the Total Environment 625:1606–1614. https://doi.org/10.1016/j.scitotenv.2017.12.336

Barrett, P.M., Hull, E.A., Burkart, K., Hargrave, O., McLean, J., Taylor, V.F., Jackson, B.P., Gawel, J.E., and Neumann, R.B. 2019. Contrasting arsenic cycling in strongly and weakly stratified contaminated lakes: Evidence for temperature control on sediment-water arsenic fluxes. Limnology and Oceanography 64(3):1333–1346. https://doi.org/10.1002/lno.11119

Blake, E. 2021. Diavik hopes to breathe new life into Frame Lake. Cabin Radio, 4 March. https://cabinradio.ca/57402/news/yellowknife/diavik-hopes-to-breathe-new-life-into-frame-lake/

Blake, E. 2022. Canada's top five federal contaminated sites to cost taxpayers billions to clean up. The Canadian Press, 28 November. https://www.theglobeandmail.com/canada/article-canadas-top-five-federal-contaminated-sites-to-cost-taxpayers-438/

Cai, M. 2005. Dynamical amplification of polar warming. Geophysical Research Letters 32(22): L22710. https://doi.org/10.1029/2005GL024481

Caumette, G., Koch, I., Estrada, E., and Reimer, K.J. 2011. Arsenic speciation in plankton organisms from contaminated lakes: Transformations at the base of the freshwater food chain. Environmental Science and Technology 45(23):9917–9923. https://doi.org/10.1021/es2025092

Caumette, G., Koch, I., House, K., and Reimer, K.J. 2014. Arsenic cycling in freshwater phytoplankton and zooplankton cultures. Environmental Chemistry 11(5):496–505. https://doi.org/10.1071/EN14039

CCME (Canadian Council of Ministers of the Environment). 2001. Canadian water quality guidelines for the protection of aquatic life: Summary table. Canadian environmental quality guidelines, 1999 (updated). Winnipeg, Manitoba: Canadian Council of Ministers of the Environment. https://ccme.ca/en/res/arsenic-en-canadian-water-quality-guidelines-for-the-protection-of-aquatic-life.pdf

Chen, G., Shi, H., Tao, J., Chen, L., Liu, Y., Lei, G., Liu, X., and Smol, J.P. 2015. Industrial arsenic contamination causes catastrophic changes in freshwater ecosystems. Scientific Reports 5: 17419. https://doi.org/10.1038/srep17419

Cohen, S. 2022. Cost of cleaning up Yellowknife's Giant Mine now pegged at $4.38B, up from $1B. CBC News, 10 November. https://www.cbc.ca/news/canada/north/giant-mine-remediation-cost-4-billion-1.6647952

Cullen, W.R., and Reimer, K.J. 1989. Arsenic speciation in the environment. Chemical Reviews 89(4):713–764. https://doi.org/10.1021/cr00094a002

Cullen, W.R., Harrison, L.G., Li, H., and Hewitt, G. 1994. Bioaccumulation and excretion of arsenic compounds by a marine unicellular alga, *Polyphysa peniculus*. Applied Organometallic Chemistry 8(4):313–324. https://doi.org/10.1002/aoc.590080406

Galloway, J.M., Palmer, M., Jamieson, H.E., Patterson, R.T., Nasser, N., Falck, H., Macumber, A.L., et al. 2015. Geochemistry of lakes across ecozones in the Northwest Territories and implications for the distribution of arsenic in the Yellowknife region. Part 1: Sediments. Open File 7908. Natural Resources Canada, Geological Survey of Canada. https://doi.org/10.4095/296954

Glew, J.R. 1988. A portable extruding device for close interval sectioning of unconsolidated core samples. Journal of Paleolimnology 1(3):235–239. https://doi.org/10.1007/BF00177769

Hellweger, F.L., Farley, K.J., Lall, U., and Di Toro, D.M. 2003. Greedy algae reduce arsenate. Limnology and Oceanography 48(6):2275–2288. https://doi.org/10.4319/lo.2003.48.6.2275

Hirano, S., Kobayashi, Y., Cui, X., Kanno, S., Hayakawa, T., and Shraim, A. 2004. The accumulation and toxicity of methylated arsenicals in endothelial cells: Important roles of thiol compounds. Toxicology and Applied Pharmacology 198(3):458–467. https://doi.org/10.1016/j.taap.2003.10.023

Hocking, D., Kuchar, P., Plambeck, J.A., and Smith, R.A. 1978. The impact of gold smelter emissions on vegetation and soils of a sub-Arctic forest-tundra transition ecosystem. Journal of the Air Pollution Control Association 28(2):133–137. https://doi.org/10.1080/00022470.1978.10470580

Houben, A.J., D'Onofrio, R., Kokelj, S.V., and Blais, J.M. 2016. Factors affecting elevated arsenic and methyl mercury concentrations in small shield lakes surrounding gold mines near the Yellowknife, NT, (Canada) region. PLoS ONE 11(4): e0150960. https://doi.org/10.1371/journal.pone.0150960

Hutchinson, T.C., Aufreiter, S., and Hancock, R.G.V. 1982. Arsenic pollution in the Yellowknife area from gold smelter activities. Journal of Radioanalytical Chemistry 71(1-2):59–73. https://doi.org/10.1007/BF02516141

Jasiak, I., Wiklund, J.A., Leclerc, E., Telford, J.V., Couture, R.M., Venkiteswaran, J.J., Hall, R.I., and Wolfe, B.B. 2021. Evaluating spatiotemporal patterns of arsenic, antimony, and lead deposition from legacy gold mine emissions using lake sediment records. Applied Geochemistry 134: 105053. https://doi.org/10.1016/j.apgeochem.2021.105053

Levy, J.L., Stauber, J.L., Adams, M.S., Maher, W.A., Kirby, J.K., and Jolley, D.F. 2005. Toxicity, biotransformation, and mode of action of arsenic in two freshwater microalgae (*Chlorella* sp. and *Monoraphidium arcuatum*). Environmental Toxicology and Chemistry 24(10):2630–2639. https://doi.org/10.1897/04-580R.1

MacDonald, R.W., Harner, T., and Fyfe, J. 2005. Recent climate change in the Arctic and its impact on contaminant pathways and interpretation of temporal trend data. Science of the Total Environment 342(1-3):5–86. https://doi.org/10.1016/j.scitotenv.2004.12.059

Navratilova, J., Raber, G., Fisher, S.J., and Francesconi, K.A. 2011. Arsenic cycling in marine systems: Degradation of arsenosugars to arsenate in decomposing algae, and preliminary evidence for the formation of recalcitrant arsenic. Environmental Chemistry 8(1):44–51.
https://doi.org/10.1071/EN10107

Palmer, M.J., Galloway, J.M., Jamieson, H.E., Patterson, R.T., Falck, H., and Kokelj, S.V. 2016. The concentration of arsenic in lake waters of the Yellowknife area. NWT Open File 2015-06. Yellowknife, Northwest Territories: Northwest Territories Geological Survey.
https://doi.org/10.46887/2015-06

Palmer, M.J., Chételat, J., Richardson, M., Jamieson, H.E., and Galloway, J.M. 2019. Seasonal variation of arsenic and antimony in surface waters of small subarctic lakes impacted by legacy mining pollution near Yellowknife, NT, Canada. Science of the Total Environment 684:326–339.
https://doi.org/10.1016/j.scitotenv.2019.05.258

Persaud, A.A., Cheney, C.L., Sivarajah, B., Blais, J.M., Smol, J.P., and Korosi, J.B. 2021. Regional changes in Cladocera (Branchiopoda, Crustacea) assemblages in subarctic (Yellowknife, Northwest Territories, Canada) lakes impacted by historic gold mining activities. Hydrobiologia 848(6):1367–1389.
https://doi.org/10.1007/s10750-021-04534-9

Pienitz, R., Smol, J.P., and MacDonald, G.M. 1999. Paleolimnological reconstruction of Holocene climatic trends from two boreal treeline lakes, Northwest Territories, Canada. Arctic, Antarctic, and Alpine Research 31(1):82–93.
https://doi.org/10.1080/15230430.1999.12003283

Pothier, M.P., Lenoble, V., Garnier, C., Misson, B., Rentmeister, C., and Poulain, A.J. 2020. Dissolved organic matter controls of arsenic bioavailability to bacteria. Science of the Total Environment 716: 137118.
https://doi.org/10.1016/j.scitotenv.2020.137118

Price, A., Maher, W., Kirby, J., Krikowa, F., Duncan, E., Taylor, A., and Potts, J. 2012. Distribution of arsenic species in an open seagrass ecosystem: Relationship to trophic groups, habitats and feeding zones. Environmental Chemistry 9(1):77–88.
https://doi.org/10.1071/EN11105

Rahman, M.A., and Hasegawa, H. 2012. Arsenic in freshwater systems: Influence of eutrophication on occurrence, distribution, speciation, and bioaccumulation. Applied Geochemistry 27(1):304–314.
https://doi.org/10.1016/j.apgeochem.2011.09.020

Rahman, M.A., Hogan, B. Duncan, E., Doyle, C., Krassoi, R., Rahman, M.M., Naidu, R., Lim, R.P., Maher, W., and Hassler, C. 2014. Toxicity of arsenic species to three freshwater organisms and biotransformation of inorganic arsenic by freshwater phytoplankton (*Chlorella* sp. CE-35). Ecotoxicology and Environmental Safety 106:126–135.
https://doi.org/10.1016/j.ecoenv.2014.03.004

Ratkowsky, D.A., Olley, J., McMeekin, T.A., and Ball, A. 1982. Relationship between temperature and growth rate of bacterial cultures. Journal of Bacteriology 149(1):1–5.
https://doi.org/10.1128/jb.149.1.1-5.1982

Rodie, A., Gerits, J.J.P., and Azcue, J.M. 1995. Biogeochemical pathways of arsenic in lakes. Environmental Reviews 3(3-4):304–317.
https://doi.org/10.1139/a95-016

Sivarajah, B., Korosi, J.B., Blais, J.M., and Smol, J.P. 2019. Multiple environmental variables influence diatom assemblages across an arsenic gradient in 33 subarctic lakes near abandoned gold mines. Hydrobiologia 841(1):133–151.
https://doi.org/10.1007/s10750-019-04014-1

Sivarajah, B., Cheney, C.L., Perrett, M., Kimpe, L.E., Blais, J.M., and Smol, J.P. 2020. Regional gold mining activities and recent climate warming alter diatom assemblages in deep sub-Arctic lakes. Polar Biology 43:305–317.
https://doi.org/10.1007/s00300-020-02635-0

Sivarajah, B., Simmatis, B., Favot, E.J., Palmer, M.J., and Smol, J.P. 2021. Eutrophication and climatic changes lead to unprecedented cyanobacterial blooms in a Canadian sub-Arctic landscape. Harmful Algae 105: 102036.
https://doi.org/10.1016/j.hal.2021.102036

Sommer, U., Gliwicz, Z.M., Lampert, W., and Duncan, A. 1986. The PEG*-model of seasonal succession of planktonic events in fresh waters. Archiv für Hydrobiologie 106(4):433–471.
https://doi.org/10.1127/archiv-hydrobiol/106/1986/433

Sommer, U., Adrian, R., De Senerpont Domis, L., Elser, J.J., Gaedke, U., Ibelings, B., Jeppesen, E., et al. 2012. Beyond the plankton ecology group (PEG) model: Mechanisms driving plankton succession. Annual Review of Ecology, Evolution, and Systematics 43:429–448.
https://doi.org/10.1146/annurev-ecolsys-110411-160251

Stewart, E.M., Hargan, K.E., Sivarajah, B., Kimpe, L.E., Blais, J.M., and Smol, J.P. 2018. A paleoenvironmental study tracking eutrophication, mining pollution, and climate change in Niven Lake, the first sewage lagoon of Yellowknife (Northwest Territories). Arctic 71(2):201–217.
https://doi.org/10.14430/arctic4720

Sweetman, J.N., and Smol, J.P. 2006. Reconstructing fish populations using *Chaoborus* (Diptera: Chaoboridae) remains – A review. Quaternary Science Reviews 25(15-16):2013–2023. https://doi.org/10.1016/j.quascirev.2006.01.007

Tenkouano, G.-T., Cumming, B.F., and Jamieson, H.E. 2019. Geochemical and ecological changes within Moira Lake (Ontario, Canada): A legacy of industrial contamination and remediation. Environmental Pollution 247:980–988. https://doi.org/10.1016/j.envpol.2019.01.019

Thienpont, J.R., Korosi, J.B., Hargan, K.E., Williams, T., Eickmeyer, D.C., Kimpe, L.E., Palmer, M.J., Smol, J.P., and Blais, J.M. 2016. Multi-trophic level response to extreme metal contamination from gold mining in a subarctic lake. Proceedings of the Royal Society B: Biological Sciences 283(1836): 20161125. http://doi.org/10.1098/rspb.2016.1125

Vaquer-Sunyer, R., and Duarte, C.M. 2008. Thresholds of hypoxia for marine biodiversity. Proceedings of the National Academy of Sciences 105(40):15452–15457. https://doi.org/10.1073/pnas.0803833105

Vucic, J.M., Gray, D.K., Cohen, R.S., Syed, M., Murdoch, A.D., and Sharma, S. 2020. Changes in water quality related to permafrost thaw may significantly impact zooplankton in small Arctic lakes. Ecological Applications 30(8): e02186. https://doi.org/10.1002/eap.2186

Wagemann, R., Snow, N.B., Rosenberg, D.M., and Lutz, A. 1978. Arsenic in sediments, water and aquatic biota from lakes in the vicinity of Yellowknife, Northwest Territories, Canada. Archives of Environmental Contamination and Toxicology 7:169–191. https://doi.org/10.1007/BF02332047

Walker, S.R., Jamieson, H.E., Lanzirotti, A., Hall, G.E.M., and Peterson, R.C. 2015. The effect of ore roasting on arsenic oxidation state and solid phase speciation in gold mine tailings. Geochemistry: Exploration, Environment, Analysis 15(4):273–291. https://doi.org/10.1144/geochem2013-238

Wang, S., and Mulligan, C.N. 2006. Occurrence of arsenic contamination in Canada: Sources, behavior and distribution. Science of the Total Environment 366(2-3):701–721. https://doi.org/10.1016/j.scitotenv.2005.09.005

Wang, Y., Wang, S., Xu, P., Liu, C., Liu, M., Wang, Y., Wang, C., Zhang, C., and Ge, Y. 2015. Review of arsenic speciation, toxicity and metabolism in microalgae. Reviews in Environmental Science and Bio/Technology 14:427–451. https://doi.org/10.1007/s11157-015-9371-9

Weber, F.-A., Hofacker, A.F., Voegelin, A., and Kretzschmar, R. 2010. Temperature dependence and coupling of iron and arsenic reduction and release during flooding of a contaminated soil. Environmental Science and Technology 44(1):116–122. https://doi.org/10.1021/es902100h

WHO (World Health Organization). 2017. Guidelines for drinking-water, 4th ed., incorporating the first addendum. Geneva. https://www.who.int/publications/i/item/9789241549950

Wrye, L.A. 2008. Distinguishing between natural and anthropogenic sources of arsenic in soils from the Giant Mine, Northwest Territories and the North Brookfield Mine, Nova Scotia. MSc thesis, Queen's University, Kingston, Ontario, Canada. https://qspace.library.queensu.ca/server/api/core/bitstreams/328f4bc6-f2f8-4ba4-af7a-90fdfcd993fa/content

Zhang, J., Ni, Y., Ding, T., and Zhang, C. 2014. The role of humic acid in the toxicity of arsenite to the diatom *Navicula* sp. Environmental Science and Pollution Research 21:4366–4375. https://doi.org/10.1007/s11356-013-2413-3

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