

Lacustrine Arcellinina (Testate Amoebae) as Bioindicators of Arsenic Contamination

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Received: 30 August 2015 / Accepted: 7 March 2016
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Abstract Arcellinids (testate amoebae) were examined from 61 surface sediment samples collected from 59 lakes in the vicinity of former gold mines, notably Giant Mine, near Yellowknife, Northwest Territories, Canada to determine their utility as bioindicators of arsenic (As), which occurs both as a byproduct of gold extraction at mines in the area and ore-bearing outcrops. Cluster analysis (Q-R-mode) and detrended correspondence analysis (DCA) reveal five arcellinid assemblages, three of which are related to varying As concentrations in the sediment samples. Redundancy analysis (RDA) showed that 14 statistically significant environmental parameters explained 57 % of the variation in faunal distribution, while partial RDA indicated that As

had the greatest influence on assemblage variance (10.7 %; $p < 0.10$). Stress-indicating species (primarily centropyxids) characterized the faunas of samples with high As concentrations (median = 121.7 ppm, max > 10000 ppm, min = 16.1 ppm, $n = 32$), while difflugiid dominated assemblages were prevalent in substrates with relatively low As concentrations (median = 30.2 ppm, max = 905.2 ppm, min = 6.3 ppm, $n = 20$). Most of the lakes with very high As levels are located downwind (N and W) of the former Giant Mine roaster stack where refractory ore was roasted and substantial quantities of As were released (as As_2O_3) to the atmosphere in the first decade of mining. This spatial pattern suggests that a significant proportion of the observed As, in at least these lakes, are industrially derived. The results of this study highlight the sensitivity of Arcellinina to As and confirm that the group has considerable potential for assessing the impact of As contamination on lakes.

Electronic supplementary material The online version of this article (doi:10.1007/s00248-016-0752-6) contains supplementary material, which is available to authorized users.

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Keywords Arcellinina · Arsenic · Contamination · Gold mine · Multivariate analysis · Northwest Territories

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Introduction

Arcellinina (or testate amoebae) are a cosmopolitan group of benthic protists with high preservation potential that occur worldwide from the tropics to the Arctic region [1, 2] in various aquatic environments ranging from fresh to brackish water habitats [3–7]. Although preserved specimens are most common in Quaternary deposits, the arcellinid fossil record extends through the Phanerozoic [8] and into the Neoproterozoic [9]. Their soft amoeboid cell is protected by a beret- or sac-like test (shell) that

ranges in size from 5 to 300 μm and is either secreted by the organism (autogenous test) or more commonly xenogenous (built by agglutinating foreign materials, such as sand grains and diatoms frustules [10]).

Three decades of research on Arcellinina has demonstrated their considerable value as bioindicators for variable environmental parameters, including water table fluctuations [11]; lake acidity [12], land-use change [13], including that associated with metal mining [14]; water quality [15]; ecosystem health and seasonal environmental changes [16]; nutrient loading [17]; and pH variability [18]. The paleontological and paleolimnological value of the group is attributed to (1) their abundance in organic-rich surface sediments (between 500–3000 specimens per ml; [6]); (2) the resistance of their tests to dissolution; (3) rapid generation time; and (4) their sensitivity to a wide variety of environmental variables [3, 6, 13, 19–22].

Recent studies in Canada and Europe have demonstrated that distinct arcellinid assemblages, species, and strains are significantly impacted by industrial pollutants [10, 16, 23–28]. In addition, some of these studies have identified a positive correlation between particular Arcellinina strains and metalloid (e.g., Arsenic (As)) and heavy metal contamination (e.g., mercury (Hg) and lead (Pb)); [23, 27, 28]). Arcellinids are characterized by a rapid reproduction rate of days to weeks, making them particularly useful for monitoring the ecosystem health of contaminated lakes and for assessing the efficacy and progress of remediation efforts [16–18, 27, 28].

The main objectives of this study were to (1) quantify the spatial response of lacustrine Arcellinina communities and individual taxa to varying As levels in sediment-water interface samples from lakes in the Yellowknife region; (2) distinguish impacts of As contamination from other environmental controls; and (3) determine the potential of using these benthic microorganisms as an efficient and cost effective bioindicator of As contamination. The region in the vicinity of the City of Yellowknife, Northwest Territories, Canada was chosen for this research because concerns remain regarding the historic legacy of As contamination derived from the operations of several former gold mines in the region, especially Giant Mine (1948 to 2004). Shear zone gold mineralization in the Slave Geological Province, particularly in the Yellowknife Greenstone Belt, includes rocks known to contain elevated levels of metals of environmental concern (e.g., aluminum, cadmium, copper, mercury and As), which can enter surface waters in the region through physical and chemical weathering. In these rocks, As concentration is generally about 10 ppm [29–32], but can range up to 1900 ppm in tills overlying mineralized zones [33]. The region was also chosen due to the abundance of lakes in low relief topography, which provide an ideal study area to examine the spatial distribution of faunal responses (e.g., 30-km radius of Yellowknife).

Background

Arsenic Contamination from Mining, Ore Processing, and Natural Sources

The discovery of significant gold mineralization in the Yellowknife Greenstone Belt in the early 1930s led to the establishment of two major gold mines in the vicinity of the City of Yellowknife: Con Mine and Giant Mine (Fig. 1). Of the two mines, Giant Mine went on to become one of the most productive and longest running mining operations in Canadian mining history, producing about 220 t of gold from 1948 to 2004 [35, 36]. This massive production resulted in a post-World War II economic boom, which led to the establishment of Yellowknife as the capital of the Northwest Territories [37]. At the Giant Mine, most gold mineralization was associated with sulfide minerals, predominantly arsenopyrite (FeAsS). Cyanide leaching is usually used to extract gold from ore, but due to the refractory mineralogy of the gold-bearing ores in the Yellowknife Greenstone Belt, roasting was necessary to liberate the gold from the gold-bearing sulfides. Ore from the Giant Mine was roasted at a relatively low temperature (500 $^{\circ}\text{C}$) to volatilize As and antimony (Sb), thus transforming sulfide minerals into porous iron oxides of maghemite that were amenable to cyanidation [38]. A byproduct of the roasting process was a continuous release of As (predominantly arsenic trioxide (As_2O_3)) particulate and SO_x vapor directly to the atmosphere via the roaster stack. During the first decade of ore processing at Giant Mine, thousands of tons of As_2O_3 was emitted to the atmosphere (2600 t/year; [39, 40]) due to inefficient extraction practices and permissive emission control policies. Emissions were slightly reduced when the first gas cleaning technologies were introduced to the roaster in 1951 [41]. More stringent controls developed and implemented after 1958 subsequently decreased emissions, substantially reducing release of As_2O_3 to approximately 5.7 t/year and led to the eventual storage of 237,176 t of As_2O_3 within the Giant Mine complex [36, 39, 42]. However, before these significant changes were made in waste handling, more than 20,000 t of As_2O_3 was released to the environment through aerial emissions, with much of it ending up in local watersheds and deposited in local lakes [36, 41].

Arsenic may also enter surface waters in the Yellowknife region through natural weathering of bedrock, till, and other surficial materials containing elevated metal concentrations. Thus, elevated concentrations of As and other metals of concern (e.g., Al, Cd, Cu, Hg) in lake sediments in the region may be the result of natural weathering, legacy mining activities, or a combination of both [43].

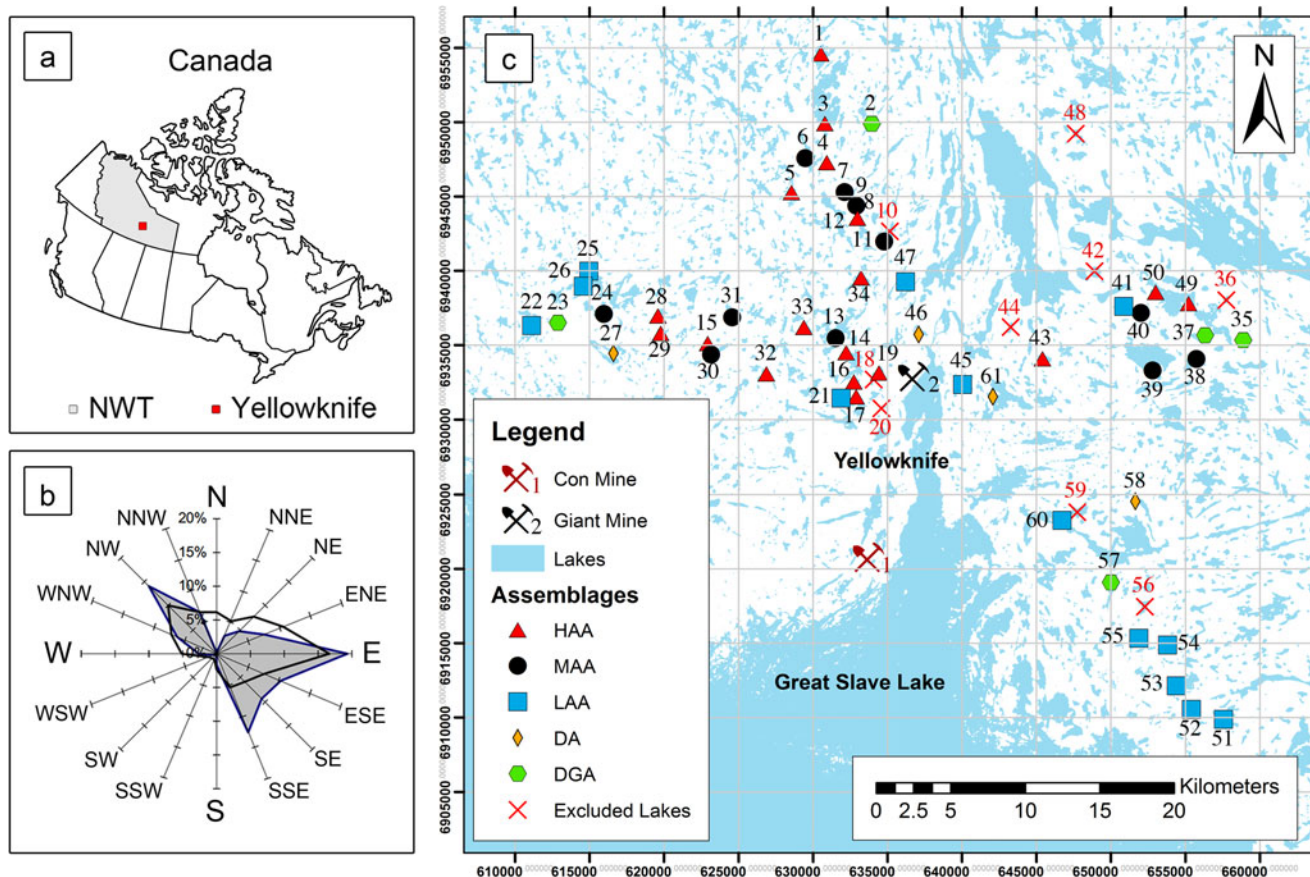


Fig. 1 Map of the study region. **a** Location of the study region in Canada. **b** Rose diagram showing the prevailing wind direction in Yellowknife (modified after Pinard et al. [34]). **c** Map of sampling sites showing the locations of the 61 sediment-water-interface samples. Abbreviations:

HAA high arsenic level assemblage, *MAA* moderate arsenic level assemblage, *LAA* low arsenic level assemblage, *DG* difflugiid assemblage, *DGA* Difflugia glans assemblage

Arsenic Bioavailability

Arsenic is a metalloid that is known to be toxic to both plants and animals, because of its affinity for protein, lipids, and other cellular components [44, 45]. Moreover, chronic exposure to As can lead to severe health effects in humans (e.g., skin lesions, anemia, liver damage, and cancer; [46]). The mobility and toxicity of As depends on its oxidation state and speciation [38, 45]. In nature, As can exist in organic or inorganic form. Inorganic forms include arsenite (AsIII) and arsenate (AsV). Organic forms include biologically methylated As compounds and monomethylarsonic acid (MMA; [45]). Arsenite is known to be more toxic than arsenate, and both are more toxic than organic As species [45, 47]. Arsenic becomes a major risk when it is bioavailable. Biological availability refers to the readiness of a chemical compound or element to be taken up by living organisms [48]. Bioavailability is influenced by many factors, such as the geology and the geochemistry of the environment, metal speciation, and residence time [49, 50]. Natural processes, such as dissolution and desorption, can also enhance the bioavailability of As by

facilitating its transformation from the solid phase to the free aqueous phase, which is more accessible for uptake by living organisms. Lake sediments offer a particularly high sorption capacity to As and can thus behave as reservoirs of As and other metals of concern that can be liberated to associated ecosystems. One of the most toxic and bioaccessible forms of As, As₂O₃ [51], is present abundantly in the lake sediments and streams in the Yellowknife area due to gold smelting associated with several mining operations, primarily the Giant Mine [36]. Previous research carried out on assessing the As levels in various mediums in the Yellowknife region suggest that current As background level in lake sediment is between ~10 to 35 ppm [52–54] and can reach to 150 ppm in soil [55]. However, these studies also show that As level can reach extreme levels in soil (500 to 9300 ppm; [56, 57]), lake sediment (1764 to 3821 ppm in the Baker Creek watershed; [58]), and surface water samples (1500 to 20,400 µg/L; [59, 60]). These levels far exceed the acceptable levels of the Canadian Council of the Ministers of the Environment (CCME) guideline for soils (12 ppm; [61]), the Government of NT guideline for industrial soils (340 ppm; [62]), the

CCME water quality guidelines for the protection of aquatic life (5 $\mu\text{g/L}$; [63]), the interim sediment quality guidelines (5.9 ppm; [61]), and the probable effect levels (17 ppm; [61]).

Remediation Plan

Although the Giant Mine has been closed since 2004, contamination remains an environmental and human health concern to the residents of Yellowknife as questions persist regarding the legacy of the enormous quantities of As_2O_3 deposited across the Yellowknife region [40, 64]. In response to this concern, the federal and territorial governments cooperated to produce the Giant Mine remediation project [65]. The remediation project has three objectives: (1) to work on stabilizing the site of the Giant Mine; (2) to isolate contamination from the surrounding environment; and (3) to rehabilitate the mine site to a safe condition in order to restore ecological processes on the mine lease area [65]. Numerous environmental studies have been conducted on the mine lease area and known discharge pathways of Giant and Con mines since the 1970s (e.g., [54, 57, 66]). However, little is known about the ecological impact of Giant and Con mines on the surrounding region ([66]; a summary of literature dealing with environmental studies is presented in Galloway et al. [67]).

Regional Setting

Lakes investigated in this study are located in the central Northwest Territories in the Yellowknife Supergroup (Fig. 1, Supplementary Table S1). These lakes occur within the Yellowknife Supergroup of the southern Slave structural province of the Canadian Shield (Fig. 1, Supplementary Table S1). Bedrock is composed of Archean metavolcanic and metasedimentary rocks, which are intruded by younger granitoids. A more detailed description of the geology of the study region is presented in [68], [69], [70], [71], [72].

Bedrock outcrops are abundant (up to 75 % of the surface) in the Yellowknife region [73]. The most prevalent surficial sediments in the study region are a mixture of Glacial Lake McConnell sediments and tills that form a thick (<2 m thick) discontinuous veneer [73]. Till consists of loosely compacted, stony, matrix-supported diamicton [73]. Clasts consist of various lithologies and range in size from small pebbles to large boulders and compose 20 to 60 % of the till in the Yellowknife region [73]. Glaciofluvial sediments consist of fine sand to cobbles in the forms of eskers, kames, and outwash and are relatively uncommon in the study region [73]. A number of surficial sedimentary deposits may be attributed to Glacial Lake McConnell, which formed in Great Slave Lake, Great Bear Lake, and Athabasca Lake basins during deglaciation of the study region approximately 10,000 years ago [73, 74]. Sedimentary deposits of Glacial Lake McConnell consist of poorly to moderately sorted coarse to fine sand, silt, and clay

that can be up to 20 m thick in some topographic lows [73]. These sediments may overlie bedrock, till, outwash, or finer-grained sediments deposited in deep water environments and may be overlain by sand and gravel representing regressive fluvial or littoral successions. Accumulations of Holocene-aged peat also occur in the study region and can be 1 m thick or greater in bogs and other low-lying wetland types [73].

Elevations in the region rise gradually from 157 m above mean sea level at Great Slave Lake to 350–400 m above mean sea level north of Thistlethwaite Lake [41, 73]. The low-relief terrain surrounding the Yellowknife region consists of rocky outcrops associated with glacial and glaciolacustrine sediments in topographic lows [41, 73]. The Yellowknife River drains the region, flowing south into Yellowknife Bay, Great Slave Lake. Most streams and rivers are shallow, and few have cut into the underlying bedrock or surficial sediments. As a result, numerous small elongated lakes have formed in shallow depressions along fault lines and joints in the bedrock [67]. Yellowknife has a subarctic, continental climate characterized by short, dry, cool summers with a mean annual temperature of -4.3 °C and a low mean annual precipitation of 170.7 mm [75]. Prevailing wind direction changes throughout the year, with the dominant wind direction being out of the east and south [76].

Materials and Methods

Sampling Design and Field Methods

In August 2012, 61 surface sediment samples were collected from 59 lakes within a 30-km radius of the City of Yellowknife. The sampled lakes were broadly distributed along four ~40-km long transects (north, south, east, and west of Giant Mine; Fig. 1) to ensure coverage of lakes that spanned the maximum possible range of influence of aerial fallout from the roaster at the Giant Mine site. Lakes were accessed by using a pontoon-equipped Bell Long Ranger helicopter. Surface sediment samples were collected using an Ekman Grab sampler. The upper 5 mm of sediment from each Ekman Grab was retained for arcellinid, sedimentological, and geochemical analysis. The onboard helicopter geographic positioning system (GPS) was used to record the location of each station (Supplementary Table S1). Sampling depth was determined for each station using a commercial “fish finder” (Lowrance Elite-4 \times) coupled with a bottom sensor indicator. Muddy substrates were preferentially selected for sampling, as nutrient-poor silt to sand substrates are generally characterized by limited arcellinid populations [6]. Surface water samples were also collected at each station to determine the concentration of nutrients in the water column. A YSI Professional Plus handheld multiparameter instrument equipped with quart cables was used to record water property data including pH,

temperature (°C), conductivity (μs), and dissolved oxygen (DO mg/l) at 1-m depth intervals through the water column and at the sediment-water interface (Supplementary Table S2).

Laboratory Methods

Particle Size Analysis

Sediment subsamples for particle size analysis were digested in a heated bath (50 °C) with 10 % HCl and 35 % H₂O₂ to remove carbonate and organics, respectively [8, 77]. Digested samples were analyzed using a Beckman Coulter LS 13 320 laser diffraction analyzer fitted with a universal liquid medium (ULM) sample chamber over a measurement range of between 0.4 and 2000 μm. The samples were loaded into the instrument until an obscuration level of 10 ± 3 % was attained. GRADISTAT (Version 8; [78]) was used to compile the results (Supplementary Table S3).

Geochemical Analysis

Trace element concentrations of lake sediment subsamples were analyzed at ACME Analytical Laboratories Ltd (Vancouver; ICP-MS 1 F/AQ250 package) by Ultratrace inductively coupled plasma mass spectrometry (ICP-MS; Supplementary Table S4). Aqua regia digestion protocol (HNO₃/HCl, 1:3) was used to extract metals that could become bioavailable (i.e., are not contained within mineral matrices). Surface water samples were analyzed for nutrients by Caduceon Environmental Laboratories, Ottawa, for nitrate (mg/l), ammonia (mg/l), total Kjeldahl nitrogen (mg/l), and total phosphorous (TP; mg/l).

Rock Eval. Pyrolysis

The type and quantity of organic matter in each lake sediment sample was determined by thermal devolatilization of organic constituents using Rock-Eval[®] 6 Analysis (Vinci Technologies, Rueil-Malmaison, France; [79]). Rock-Eval[®] 6 Analysis uses heat to break down large organic matter molecules to smaller and chemically more identifiable molecules [80]. Quantitative measurements of total organic carbon (TOC) and other organic geochemical parameters, including S1, S2, and S3, were produced (Supplementary Table S5). S1 carbon represents the quantity of free hydrocarbons in sediments (mg hydrocarbons/g) that are devolatilized during pyrolysis at 300 °C. S2 carbon represents the quantity of large molecules, kerogen-derived hydrocarbons released through thermal cracking of the organic matter, in sediment samples (mg hydrocarbons/g) near 650 °C. The S2 compounds in sediment generally correspond to highly aliphatic biomacromolecule structures of algal cell walls [81]. S3 represents the amount of carbon dioxide released during pyrolysis of kerogen. The quantity of all organic matter

released during pyrolysis and oxidation heating accounts for TOC (wt.%) in sediment samples. Analyses of standard reference material (IFP 160000, Institut Français du Pétrole and internal 9107 shale standard, Geological Survey of Canada, Calgary) show accuracy and precision to be greater than 5 % RSD.

Micropaleontological Analysis

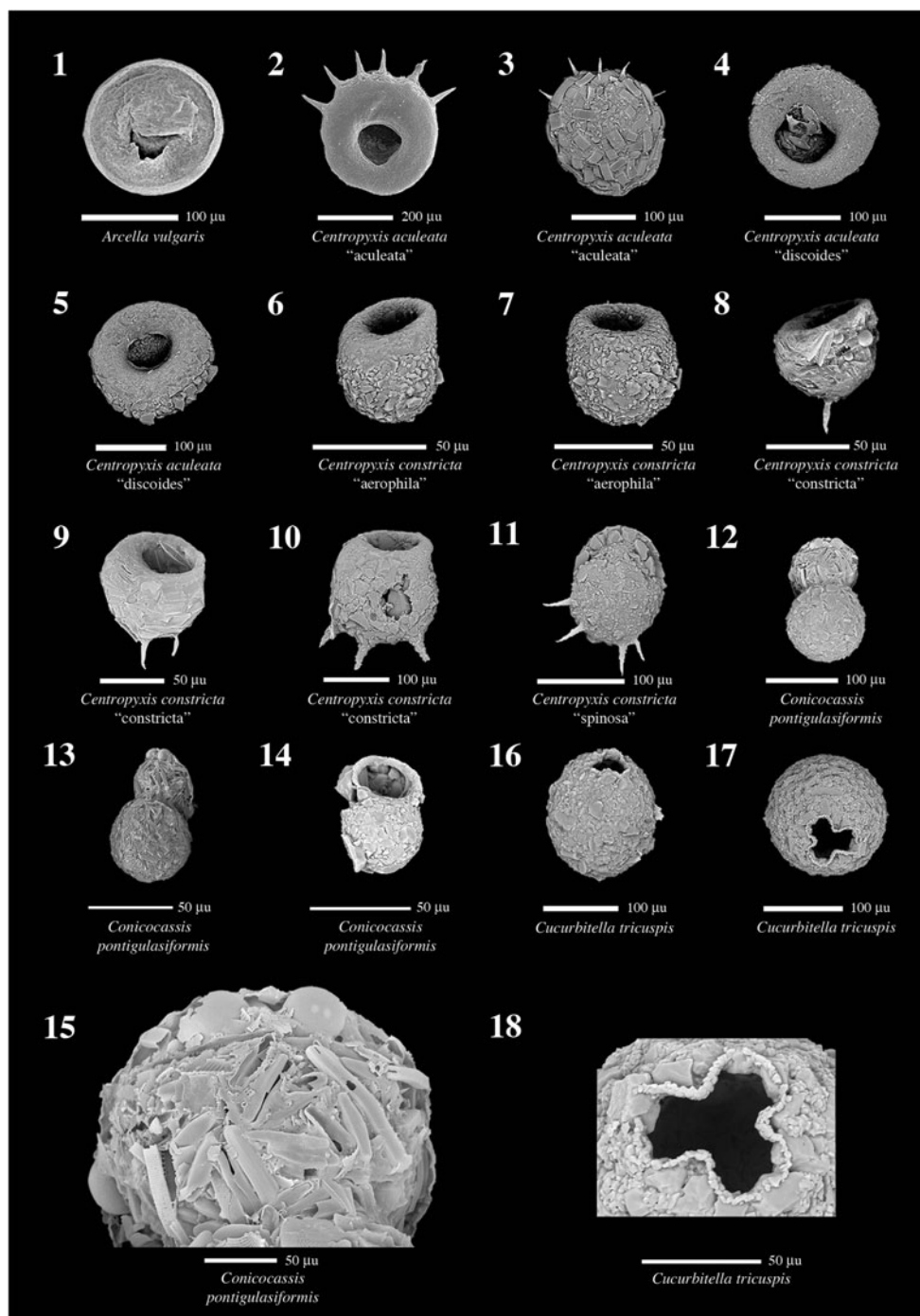
Separate sediment subsamples (2.5 cm³) were used for micropaleontological analysis. These samples were wet sieved through a 297-μm mesh to remove any coarse debris (e.g., grass and sticks) and then through a 37-μm mesh to separate arcellinids from the clay fraction. Samples were preserved with isopropyl alcohol and subdivided into six aliquots for quantitative analysis using a wet-splitter (after Scott and Hermelin [82]). Aliquots were quantitatively analyzed wet for the contained arcellinids on a gridded Petri dish using an Olympus SZH dissecting binocular microscope (×7.5–64 magnification) until, whenever possible, a statistically significant number of specimens were quantified (Supplementary Table S7; [83]). Three samples (BC 10, 20, and 48) had statistically insignificant numbers of arcellinid tests and were thus removed from ensuing statistical analysis.

Identification of arcellinids primarily followed the illustrations and descriptions found in various key papers where specimens are well illustrated (e.g., [5, 15, 17, 18, 28]). To circumvent taxonomic issues associated with the prevalence of phenotypic plasticity in Arcellinina, phenotypes characterized by stable morphologies are designated as “strains” (after [23, 28, 84]). Strains are not valid taxonomic divisions according to the International Code of Zoological Nomenclature [85], but as many of these infrasubspecific morphologies have been demonstrated to be ecophenotypes, their use increases the utility of using Arcellinina as environmental indicators [10, 23, 24, 27, 84]. Scanning electron microscope images of common species and strains were obtained using a Tescan Vega-II XMU VP scanning electron microscope (SEM) in the Carleton University Nano Imaging Facility. All SEM plates were digitally produced using Adobe Photoshop[™] CS12 on an Apple Macintosh[®] computer (Figs. 2 and 3).

Statistical Analysis

Twenty-nine arcellinid species and strains were identified in the 61 lake sediment subsamples. The Probable Error (pe) was calculated for each sample [83]. A sample count was deemed statistically insignificant if the probable error exceeded the total count for a sample. Three samples (BC 10, 20, and 48) contained statistically insignificant populations and were thus excluded from the subsequent multivariate data analyses. Standard error (Sxi) was also calculated for each sample [83]. Species was considered to be present in insignificant

Fig. 2 Scanning electron microscope of selected arcellinid shells from the study lakes. For more specimen information, see SA 7. *1 Arcella vulgaris* Ehrenberg 1830 specimen from sample BC 54. *2–3 Centropyxis aculeata* (Ehrenberg 1832) stain “aculeata” for specimens from sample BC 44. *4–5 Centropyxis aculeata* (Ehrenberg 1832) stain “discoides” for specimens from samples BC 9 and BC 30. *6–7 Centropyxis constricta* (Ehrenberg 1843) stain “aerophila” specimens from samples BC 8 and BC 17. *8–10 Centropyxis constricta* (Ehrenberg 1843) stain “constricta” specimens from samples BC 30 and BC 3. *11 Centropyxis constricta* (Ehrenberg 1843) stain “spinosa” from sample BC 11. *12–15 Coniocassiss pontigulasiformis* (Beyens et al. 1986), 2015 specimens from samples BC 25 and BC 46. *14 A* specimen from sample BC 46. *15* Details of the visor surrounding the aperture. *16–18 Cucurbitella tricuspis* (Carter 1956) specimens from samples BC 8 and 51. *18* Characteristic lobes of the aperture



number if the standard error exceeded the total counts for that species in all samples. Out of the 29 species and strains, 5 (4 species and 1 strain) were found to be present in statistically insignificant numbers. These species and strains were also excluded from the ensuing statistical analysis.

The Shannon Diversity Index (SDI; [86]) was used to examine the faunal diversity of the species found in each sample to provide a general indication of the relative health of the sampled lake. Environments are considered to be stable if

the SDI falls between 2.5 and 3.5, in transition between 1.5 and 2.5, and stressed between 0.1 and 1.5 [10, 87].

Data Screening

Micropaleontological and geochemical datasets were screened prior to statistical analysis. Following Reimann et al. [88], any variables having issues associated with more than 25 % of their values (i.e., missing values, below detection, or above

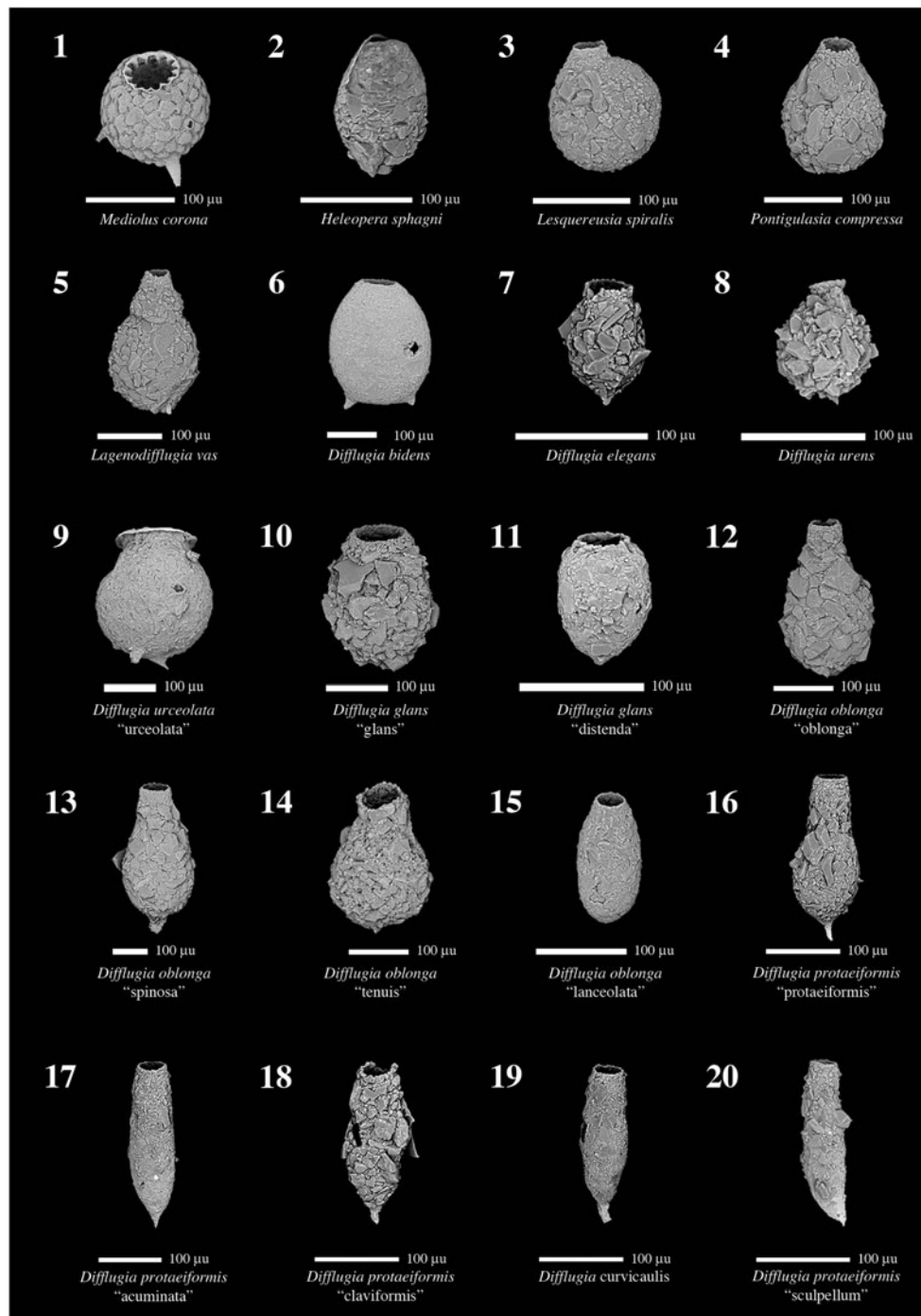


Fig. 3 Scanning electron microscope of selected arcellinid tests from the study lakes. For more specimen information see SA 7. 1 *Mediolus corona* (Wallich 1986) specimen from sample BC 51. 2 *Heleopera sphagni* (Leidy 1874) specimen from sample BC 9. 3 *Lesquereusia spiralis* (Ehrenberg 1840) specimen from sample BC52. 4 *Pontigulasia compressa* (Carter 1864) specimen from sample BC 30. 5 *Lagenodifflugia vas* (Leidy 1874) specimen from sample BC 9. 6. *Difflugia bidens* Penard 1902 from sample BC 6. 7 *Difflugia elegans* Penard, 1890. 8 *Difflugia urens* Patterson et al. 1985 specimen from sample BC 27. 9 *Difflugia urceolata* Carter 1864 strain “urceolata” specimen from sample BC 24. 10 *Difflugia glans* Penard 1902 strain “glans” specimen from sample BC 52. 11 *Difflugia glans* Penard 1902 strain “distenda” specimen from sample BC 46. 12 *Difflugia oblonga*

Ehrenberg 1832 strain “oblonga” specimen from sample BC 23. 13 *Difflugia oblonga* Ehrenberg 1832 strain “spinosa” specimen from sample BC 35. 14 *Difflugia oblonga* Ehrenberg, 1832 strain “tenuis” specimen from sample BC 39. 15 *Difflugia oblonga* Ehrenberg 1832 strain “lanceolata” specimen from samples BC 46. 16 *Difflugia protaeiformis* Lamarck 1816 strain “protaeiformis” specimen from samples BC 46. 17 *Difflugia protaeiformis* Lamarck 1816 strain “acuminata” specimen from samples BC 38. 18 *Difflugia protaeiformis* (Lamarck 1816) strain “claviformis” specimen from samples BC 52. 19 *Difflugia curvicaulis* (Penard 1899) specimen from samples BC 9. 20 *Difflugia protaeiformis* Lamarck, 1816 strain “sculpellum” specimen from samples BC 9

detection) were removed (Supplementary Table S8). This procedure screened out six samples (BC 18, 36, 42, 44, 56, and 59) from the dataset. For variables where less than 25 % of the cases were below the lower detection limit, these entries were reported as a value equal to half the detection limit. For cases where the variable was present above the upper instrumental detection limit, entries were reported as a value equal the upper detection

limit [88]. All concentration variables were converted to parts per million (ppm).

Variables Reduction

Large ecological data sets are often problematic to analyze as they may contain redundancies in environmental information

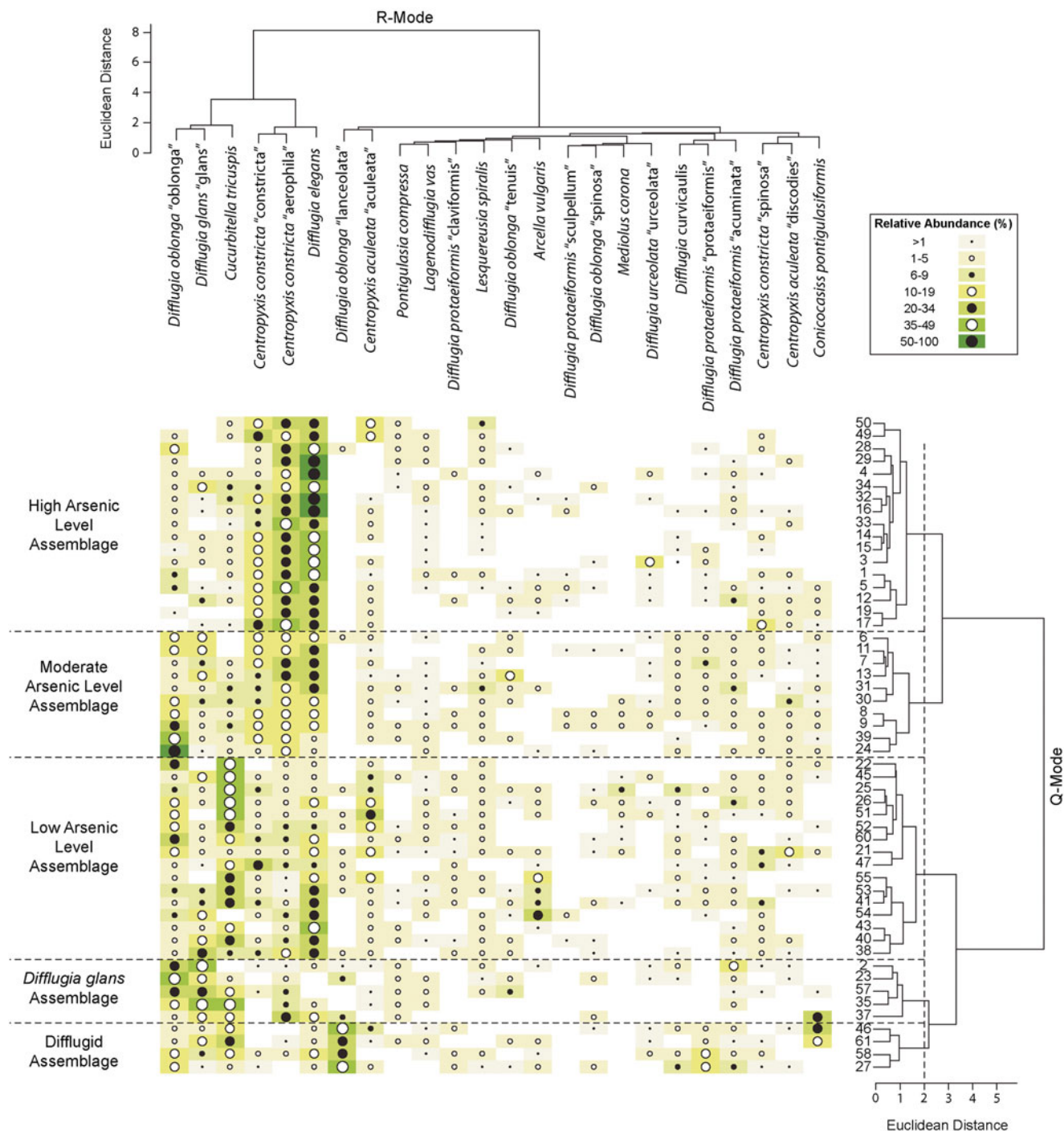


Fig. 4 Combined Q-mode and R-mode cluster dendrogram for the 52 samples and 24 statistically significant species and strains. Five faunal assemblages are indicated. The colored squares (gradient of green) and

circles (black and white) reflect the relative abundances of the arcellinid species and strains

and/or environmental variables that have no influence on the distribution of species [89]. To deal with these potential problems, a two-stage statistical test protocol was implemented to analyze the data presented here. In the first stage, highly correlated variables with no clear impact on arcellinid distribution were removed using Pearson correlation (Supplementary Table S9). In the second stage, the collinearity of each of the remaining variables was determined using the variance inflation factor (VIF), which is a component of the USDM package in the R statistical programming environment. The program removed any variables that exceeded a predetermined VIF cutoff value (Supplementary Table S10). Any variables with VIF >10 were deemed to be highly colinear and were eliminated from subsequent analysis. Although removed by the VIF procedure, S2 carbon was included in the ensuing statistical analyses as it is generally derived from algal cell walls [81] and would thus be possibly linked to lake primary production.

Cluster Analysis

Q-mode cluster analysis was used to group samples containing statistically similar arcellinid populations using Ward’s

Minimum variance method [90] and recorded as Euclidean distances (after Fishbein and Patterson [91]). Following the same method, R-mode cluster analysis was carried out to determine which species were most closely associated with each other and thus best characterized a particular assemblage (after Fishbein and Patterson [91]). Q-mode and R-mode cluster analyses were carried out on 24 arcellinid species and strains in 52 samples determined to have statistically significant counts and not missing any values in the environmental data set. The results were generated using R statistical software package and both dendrogram were organized into a two-way hierarchical dendrogram by using Adobe Illustrator™ CS12 on an Apple Macintosh® computer (Fig. 4).

Detrended Correspondence Analysis

Detrended correspondence analysis (DCA; [92]) was used to compare the similarity between identified assemblages in multidimensional space (Fig. 5). DCA revealed a gradient length of 1.9 for the species data, which represented a unimodal response (<2) in this study. A Hellinger transformation was

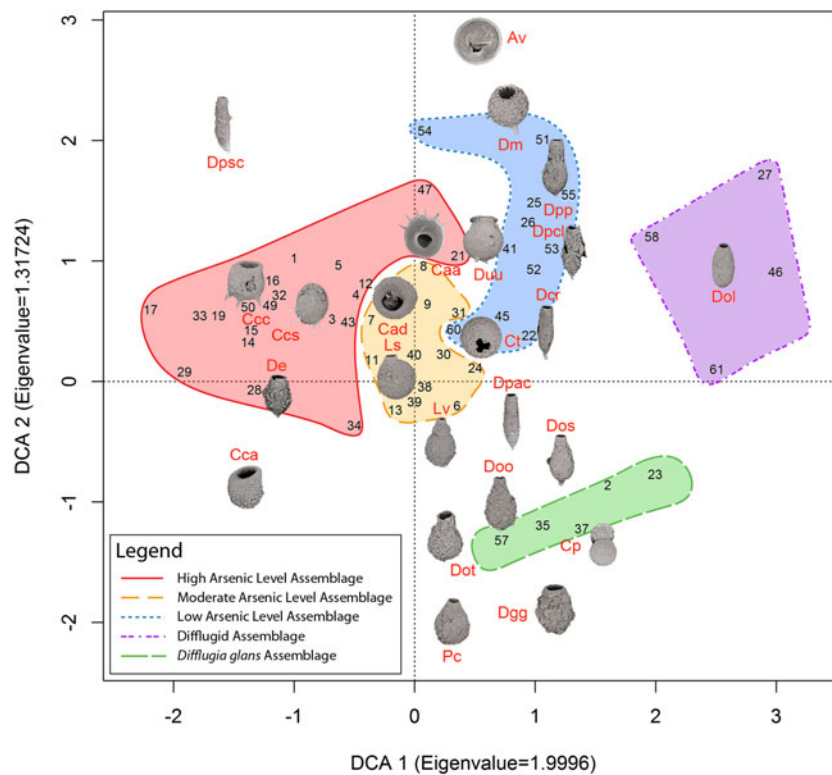


Fig. 5 Detrended Correspondence Analysis (DCA) bi-plot. Av—*Arcella vulgaris*, Caa—*Centropyxis aculeata* “aculeata,” Cad—*Centropyxis aculeata* “discoides,” Cca—*Centropyxis constricta* “aerophila,” Ccc—*Centropyxis constricta* “constricta,” Ccs—*Centropyxis constricta* “spinosa,” Cp—*Coniocassiss pontigulasiformis*, Ct—*Cucurbitella tricuspis*, Mc—*Mediolus corona*, Doo—*Diffflugia oblonga* “oblonga,” Dos—*Diffflugia oblonga* “spinosa,” Dot—*Diffflugia oblonga* “tenuis,”

Dol—*Diffflugia oblonga* “lanceolata,” Dgg—*Diffflugia glans* “glans,” Duu—*Diffflugia urceolata* “urceolata,” Dpp—*Diffflugia protaeiformis* “protaeiformis,” Dpa—*Diffflugia elegans*, Dpac—*Diffflugia protaeiformis* “acuminate,” Dpcl—*Diffflugia protaeiformis* “claviformis,” Dpccr—*Diffflugia curvicaulis* Penard, 1899, Dpsc—*Diffflugia protaeiformis* “scalpellum” Ls—*Lesquereusia spiralis*, Lv—*Lagenodifflugia vas*, Pc—*Pontigulasia compressa*

used to satisfy the assumption of the linearity made by redundancy analysis (RDA).

Redundancy Analysis

RDA [93] of the 52 samples and 24 statistically significant species and strains was used to assess the relationship between arcellinid assemblages and measured environmental variables (Fig. 6). This analysis provided important insight for interpreting the cluster analysis and DCA results. A series of partial RDAs (pRDA), coupled with the variance partitioning test, was carried out to identify the significance of the RDA axes and the measured variables. To determine the number of axes that needed to be retained, a scree plot was generated (Supplementary Fig. S1). A scree plot is a simple line segment plot that shows the fraction of total variance in the data represented by each RDA axis. The plot can also show an elbow-like

separation between significant and less significant axes. Only axes above the separation were retained. Variance partition provided an additional means of assessing the proportion of the variance in the arcellinid data set that can be attributed to the measured environmental variables (Fig. 7). Variables with a p value of <0.05 were deemed to contribute significantly to the variance in the arcellinid assemblage.

Results

Cluster Analysis

Q-mode cluster analysis of the 52 surface sediment samples retained for the study revealed 5 distinct arcellinid assemblages: (1) “High As level assemblage”; (2) “Moderate As level assemblage”; (3) “Low As level assemblage”; (4)

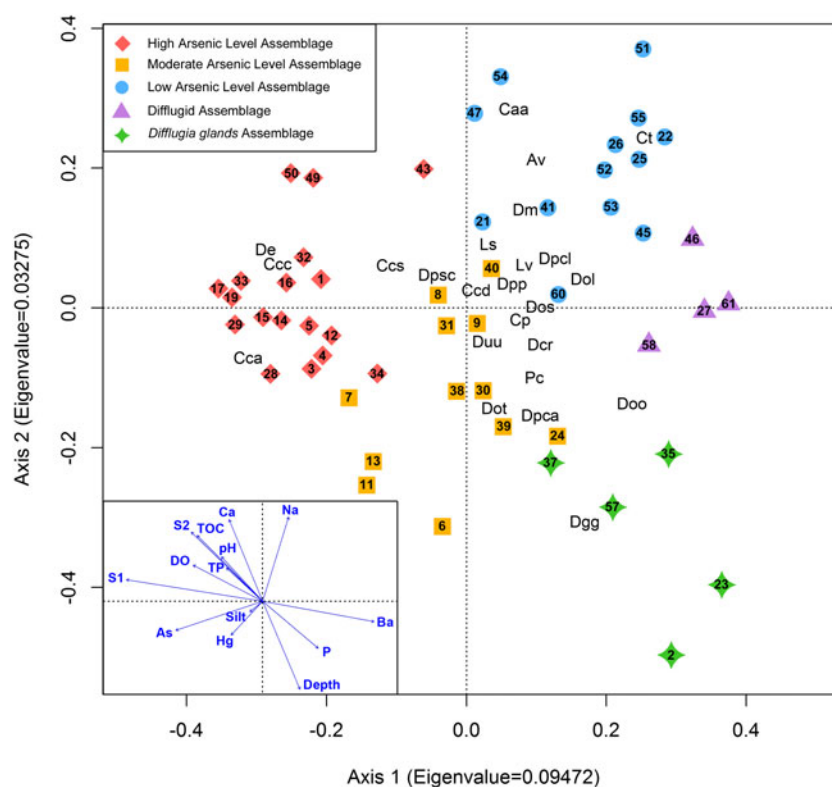
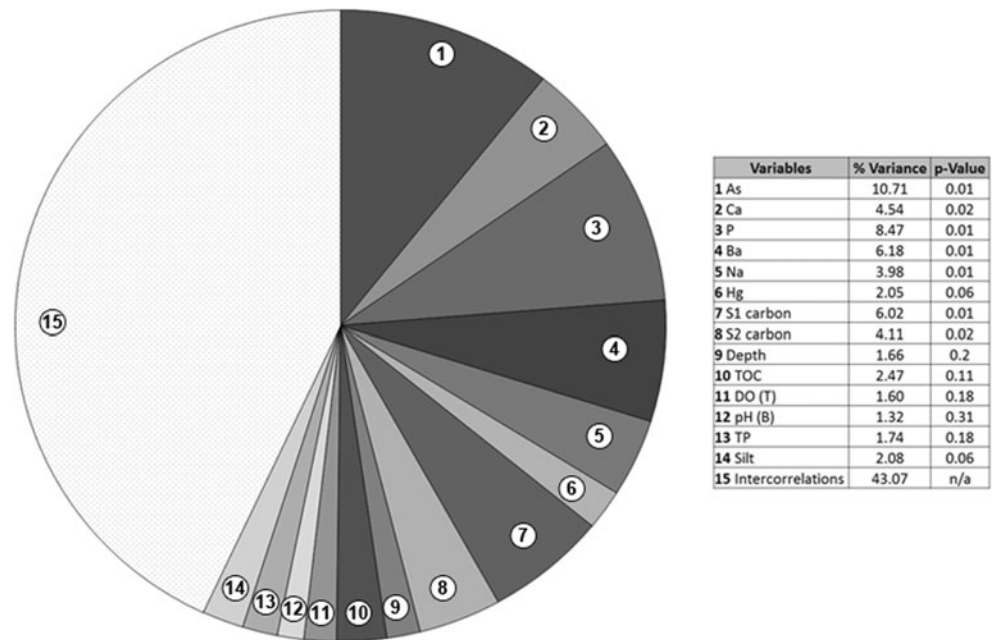


Fig. 6 Redundancy analysis (RDA) species-environment sample triplots for the 52 sediment-water interface samples that yielded statistically significant arcellinid populations and had no missing values. As—arsenic, Hg—mercury, P—sedimentary phosphorous, Ba—barium, Na—Sodium, Ca—Calcium, S1—S1 carbon, S2—S2 carbon, DO—surface dissolved oxygen, pH—bottom pH, TP—total phosphorous. Av—*Arcella vulgaris*, Caa—*Centropyxis aculeata* “aculeata,” Cad—*Centropyxis aculeata* “discoidea,” Cca—*Centropyxis constricta* “aerophila,” Ccc—*Centropyxis constricta* “constricta,” Ccs—*Centropyxis constricta* “spinosa,” Cp—*Conicocassis pontigulasiformis*,

Ct—*Cucurbitella tricuspis*, Mc—*Mediolus corona*, Doo—*Difflugia oblonga* “oblonga,” Dos—*Difflugia oblonga* “spinosa,” Dot—*Difflugia oblonga* “tenuis,” Dol—*Difflugia oblonga* “lanceolata,” Dgg—*Difflugia glans* “glans,” Duu—*Difflugia urceolata* “urceolata,” Dpp—*Difflugia protaeiformis* “protaeiformis,” Dpa—*Difflugia elegans*, Dpac—*Difflugia protaeiformis* “acuminata,” Dpcl—*Difflugia protaeiformis* “claviformis,” Dpcr—*Difflugia curvicaulis*, Dpsc—*Difflugia protaeiformis* “scalpellum,” Ls—*Lesquereusia spiralis*, Lv—*Lagenodiffugia vas*, Pc—*Pontigulasia compressa*

Fig. 7 Partial redundancy analysis (pRDA) with variance partitioning test showing the percentage variance in the arcellinid data set that is explained by the measured environmental variables and p value



“Diffflugid assemblage”; and (5) “*Diffflugia glans* assemblage” (Fig. 4). Each assemblage was named for the controlling variable, species, or environmental condition that characterized the assemblage. R-mode cluster analysis indicated that out of the 24 analyzed arcellinid species and strains, only 6 dominated the assemblage composition: *Diffflugia elegans* Penard, 1890, *Centropyxis constricta* (Ehrenberg, 1843) strain “aerophila,” *Centropyxis constricta* (Ehrenberg, 1843) strain “constricta,” *Curcurbitella tricuspis* (Carter, 1856), *Diffflugia glans* Penard, 1902 strain “glans,” and *Diffflugia oblonga* Ehrenberg, 1832 strain “oblonga” (Fig. 4).

Detrended Correspondence Analysis

Results from the DCA analysis were similar to that obtained by cluster analysis, with five distinct arcellinid assemblages also recognizable (Fig. 5). The results of the DCA analysis will be discussed in the context of the identified faunal assemblages. However, it is worth noting that cluster analysis and DCA did produce different loadings associated with samples BC 21, 38, 40, 43, and 47 (Figs. 4 and 5). Variation between these results is expected given the spatial nature of the study, which allows for a certain degree of overlap to occur between the results of these analyses.

Redundancy Analysis and Partial Redundancy Analysis

The RDA results are in general agreement with the results of the DCA and cluster analysis, as sample groupings are also characterized by the same five arcellinid assemblages (Fig. 6). Variance partition of the partial RDA results indicates

that four axes are significant at $p < 0.005$ (Supplementary Table S11). However, only the first three RDA axes were retained based on the scree plot results (Supplementary Fig. S1). RDA axes one (Eigenvalue = 0.09472), two (Eigenvalue = 0.03275), and three (Eigenvalue = 0.01501) collectively explain 42.8 % of the total variance in the arcellinid data and 75.2 % of the species-environment relationship. Variance partitioning also confirmed that 14 environmental variables influenced the faunal distribution (Fig. 7). The results indicate that As is the most statistically significant influencing variable, explaining 10.7 % of the variance within the observed arcellinid faunal distribution (Fig. 7). Other statistically significant controlling variables include sedimentary phosphorus (P; 8.5 %), barium (Ba; 6.2 %), S1 carbon (6 %), calcium (Ca; 4.5 %), and S2 carbon (4 %). All statistically significant measured environmental variables, selected through variance partitioning, collectively explained 57 % of the total variance in the arcellinid data (Fig. 7).

Arcellinid Assemblages

Assemblage 1—High As Level Assemblage (n = 20)

The faunal structure of the high As level assemblage (HAA) is dominated by *Diffflugia elegans* Penard, 1890 ($\bar{x} = 39.3 \% \pm 13$ SD), *Centropyxis constricta* (Ehrenberg 1843) strain “aerophila” ($\bar{x} = 26.7 \% \pm 9.9$ SD), and *Centropyxis constricta* (Ehrenberg 1843) strain “constricta” ($\bar{x} = 11.5 \% \pm 5.6$ SD; Fig. 4). *Diffflugia oblonga* Ehrenberg 1832 strain “oblonga” ($\bar{x} = 3 \% \pm 2.7$ SD) and *Curcurbitella tricuspis* (Carter 1856) ($\bar{x} = 3.3 \% \pm 4.6$ SD) are also common in some of the samples. HAA occurred primarily in lakes

along transects located N, W, and, to a lesser extent, E of Giant Mine (Fig. 1). The lakes are located at varying distances from Giant Mine (1.6 to 22.8 km; $\bar{x} = 11.3 \text{ m} \pm 6.7 \text{ SD}$). The samples were collected from relatively shallow sampling depths ($\bar{x} = 2.49 \text{ m} \pm 1.6 \text{ SD}$) in silt-dominated substrates ($\bar{x} = 76 \% \pm 5 \text{ SD}$). The range of observed SDI values for HAA-bearing samples (1.11–2.23) is indicative of stressed to transitional environmental conditions when anomalous sample BC 21 (SDI value 2.59) is omitted [10, 87].

The DCA results indicate that HAA-bearing samples are generally closely grouped (Fig. 5), with the exception of samples BC 21, 47, and 43. These three samples were plotted at the boundaries between assemblages 1, 2, and 3 (Fig. 5). As would be expected in any natural system, there is a gradation at the boundary between assemblages, which explains the overlapping positioning of these samples at assemblage boundaries in the DCA plot and the minor inconsistencies with the Q-mode cluster results (Fig. 4). RDA analyses show HAA correlating positively with As, as well as S1 carbon, S2 carbon, Ca, TOC and TP and negatively with depth, sedimentary P, and Ba (Fig. 6). HAA is associated with very high As concentrations ($\bar{x} = 1090.9 \text{ ppm} \pm 2477.6 \text{ SD}$) that exceed the interim sediment quality guidelines (ISQG = 5.9 ppm) and the probable effect levels (PEL = 17 ppm; [61]).

The dominant taxa in HAA, *Diffflugia elegans*, *Centropyxis constricta* strain “aerophila,” and *Centropyxis constricta* strain “constricta,” also correlated positively with As on the RDA plot (Fig. 6). *Diffflugia protaeiformis* strains are known to be closely associated with environmentally stressed, metalloid- and metal-contaminated substrates [23, 27, 28]. For example, abundant *D. elegans* has been linked to stressed environmental conditions attributed to residual mine tailings-derived contamination [28]. Likewise, centropyxid species and strains are opportunistic in nature and are capable of withstanding hostile environmental conditions, including cold temperatures [94], low salinity conditions (<5‰; [95–97]), oligotrophic conditions [98], as well as the elevated As concentrations characterizing HAA-bearing samples [27, 28].

While the abundance of stress-indicating taxa in this assemblage provides evidence of stressed environmental conditions, the low-to-moderate faunal diversity of this assemblage suggests a transition toward less hostile conditions (SDI = 1.11–2.23; [10, 87]). A similar contrast between substrate contamination levels and arcellininid diversity in Northeastern Ontario was previously considered to be an indication of ongoing remediation in stressed ecosystems [28]. Interestingly, some of HAA-bearing samples are characterized by the presence of low to moderate numbers of *D. oblonga* strain “oblonga” and *C. tricuspis* in their faunal structure. These species thrive in relatively healthy substrates rich with organics [1, 96]. Their occurrence in HAA, albeit in low to moderate abundances, suggests that natural remediation following deposition of As associated with mining activities and/

or geogenic processes may be underway. Downcore analyses are required to confirm this hypothesis as it is also possible that an intermediately stressed community has inhabited these lakes for a long period of time.

Assemblage 2—Moderate As Level Assemblage (n = 12)

Similar to HAA, the moderate As level assemblage (MAA) is dominated by stressed indicating taxa, including *D. elegans* Penard, 1890 ($\bar{x} = 19.6 \% \pm 10 \text{ SD}$), *C. constricta* (Ehrenberg 1843) strain “aerophila” ($\bar{x} = 16 \% \pm 5.7 \text{ SD}$), and *C. constricta* (Ehrenberg 1843) strain “constricta” ($\bar{x} = 9 \% \pm 3.9 \text{ SD}$; Figs. 2 and 3). The numbers of *D. oblonga* Ehrenberg 1832 strain “oblonga” ($\bar{x} = 10.4 \% \pm 18 \text{ SD}$) and *C. tricuspis* (Carter 1856) ($\bar{x} = 5.5 \% \pm 6.5 \text{ SD}$) are relatively high in comparison to HAA. *Diffflugia glans* Penard 1902 strain “glans” ($\bar{x} = 16 \% \pm 16 \text{ SD}$) is a notable additional faunal component of this assemblage. Samples with MAA are observed along transects N, E, and W, with a single sample from transect S (Fig. 1). The distance between the Giant Mine site and lakes characterized by this assemblage varied from 5.3 to 20.4 km ($\bar{x} = 14 \text{ km} \pm 4.2 \text{ SD}$). MAA was characterized by an average sampling depth of 4.7 m ($\pm 3.9 \text{ SD}$). As observed in HAA-bearing samples, sediments were dominantly silt ($\bar{x} = 68 \% \pm 18 \text{ SD}$), but with a slightly higher sand content ($\bar{x} = 18 \% \pm 21 \text{ SD}$). The SDI values derived for MAA ranged from 1.64 to 2.55 being indicative on transitional to relatively healthy environmental conditions [10, 87].

As with HAA, the DCA results for MAA samples differed slightly from what was observed in the cluster analysis. Samples BC 38 and 40 are clustering with the MAA samples in the DCA plot, while cluster analysis shows both samples in the low arsenic level assemblage (LAA). This difference is attributed to the gradational nature of the boundaries between assemblages (Figs. 4 and 5). The RDA analysis results show that MAA correlated positively with As, Hg, silt, depth, and sedimentary P and negatively with Na, Ca, TOC, pH, S2, and DO (Fig. 6). The ICP-MS analysis results indicated that samples comprising this assemblage were characterized by moderately high sediment As concentrations ($\bar{x} = 148 \text{ ppm} \pm 208 \text{ SD}$), which were notably lower than the As concentrations in HAA. However, the concentrations of As in MAA are still higher than the acceptable levels of both the ISQG and PEL [61]. Similar to HAA, the lakes supporting MAA were mainly from transects W and N. Therefore, the elevated sediment As levels associated with MAA are also consistent with the aerial emission of As_2O_3 from Giant Mine operation. However, the notable reduction in As concentration is expected as most of the MAA-bearing lakes, with the exception of samples BC 11 and 13, are located between 12 to 20 km away from mine site (Fig. 1).

The composition of the arcellininid species and strains comprising MAA is consistent with a reduced influence of

As on the assemblage relative to HAA. Stress-indicating taxa, including *D. elegans*, *C. constricta* strain “aerophila,” and *C. constricta* strain “constricta,” dominated the faunal make-up of MAA but were present in lower proportions compared to HAA. A notable change in the faunal structure of MAA is the increase in the relative abundance of *D. oblonga* “oblonga.” This arcellinid strain thrives in organic-rich sediments from tropical to Arctic conditions [1]. Although the average TOC associated with MAA does not reflect eutrophic conditions ($\bar{x} = 18.9 \pm 8.7$ SD), the high diversity of arcellinids in this assemblage suggests the presence of sufficient amounts of organics to sustain a considerable diversity of arcellinid species. The reduction in the relative abundance of stress-indicating taxa and the increased proportions of *D. oblonga* strain “oblonga,” along with the higher SDI values, provide corroborative evidence that environmental stress in MAA-bearing samples was reduced relative to the conditions observed in HAA.

Of particular note for MAA was the presence of significant proportions of *D. glans* Penard 1902 strain “glans,” which Reinhardt et al. [28] reported from relatively deep, contaminated lake substrates in Northeastern Ontario. However, the notable increase in the abundance of this strain in MAA may be more closely correlated with depth than As contamination as the RDA analysis indicates a close correlation between the distribution of this strain and sampling depth (Fig. 6).

Assemblage 3—Low As Level Assemblage ($n = 11$)

The fauna characterizing the low As level assemblage (LAA) samples was similar to those observed in HAA and MAA, being dominated by *D. elegans* Penard, 1890 ($\bar{x} = 13 \% \pm 8.5$ SD). However, a major difference from the faunal makeup of the first two assemblages is the codominance of *C. tricuspis* (Carter 1856) ($\bar{x} = 27.5 \% \pm 10.8$ SD). Centropyxids, especially *C. constricta* (Ehrenberg 1843) strain “aerophila” ($\bar{x} = 3.5 \% \pm 2$ SD), *C. constricta* (Ehrenberg 1843) strain “constricta” ($\bar{x} = 5.7 \% \pm 6.5$ SD), and *C. aculeata* (Ehrenberg 1832) strain “aculeata” ($\bar{x} = 10 \% \pm 7$ SD) are also important assemblage components present in moderate abundances. Notable numbers of *Diffugia oblonga* Ehrenberg 1832 strain “oblonga” ($\bar{x} = 10.7 \% \pm 7.8$ SD) were present in some samples. LAA-bearing samples were distributed through all transects (Fig. 1). The LAA-bearing lake closest to Giant Mine was 4 km (BC 45) from the mine site, while the furthest lake was located 31.3 km away from the mine (BC 51; $\bar{x} = 19.3 \text{ km} \pm 9.5$ SD). The lakes characterized by this assemblage were the shallowest of all lakes sampled for this study ($2 \text{ m} \pm 1.2$ SD). The substrate characterizing the stations where these samples were collected was typically silty ($\bar{x} = 70.2 \% \pm 11.7$ SD). The observed SDI values for LAA (between 1.93 and 2.47) are indicative of transitional environmental conditions [10, 87].

The DCA supports the designation of the LAA from cluster analysis, except for samples BC 21 and 47, which overlapped with HAA, and sample BC 43, which was plotted with HAA (Figs. 4 and 5). The RDA analysis results show the samples of this assemblage correlating positively with Na and negatively with As, Hg, and silt (Fig. 6). Geochemical analysis revealed that LAA, with the exception of samples BC 21 and 47, corresponded with the lowest sediment As concentrations ($\bar{x} = 26 \text{ ppm} \pm 123$ SD). Although, low As concentrations in 77 % ($n = 10$) of samples containing LAA remain well above the levels proposed by the ISQG and PEL [61]. The lakes supporting LAA are mostly upwind to Giant Mine (transects to the south and, to a lesser extent, east of the mine). This may explain the relatively low sediment As concentrations associated with LAA, as these lakes are located beyond the influence of the prevalent wind coming from the west and north of Yellowknife. Three samples, collected from lakes within the western transect, are characterized by relatively low As concentrations (Samples BC 22, 25, and 26; average As level 37.8 ppm). These lakes are among the furthest from Giant Mine in the western transect (average distance from the Giant Mine = 23 km) and thus would be expected to have relatively low As concentrations.

Compared to HAA and MAA, the arcellinid makeup of LAA is characterized by lower abundances of stress-indicator species and strains, such as *D. protaeiformis* strains and centropyxids. This is consistent with the observation that LAA-bearing samples are characterized by the lowest As concentrations in the data set when anomalous samples BC 21 and BC 47 are excluded. Furthermore, the majority of these samples were collected from lakes located at a considerable distance from Giant Mine ($\bar{x} = 19 \text{ km} \pm 9.5$ SD; Fig. 1). These lakes may, therefore, have never been significantly impacted by As aerial fallout from the mine site roaster. The negative correlation between LAA and As, shown by the RDA plot, also confirms the weak influence of As on the faunal structure of the assemblage (Fig. 6) and explains the high arcellinid diversity characterizing the assemblage. The environmental conditions in lakes bearing LAA are similar to previously observed assemblages (see the high diversity assemblage (2) of [28]) and are indicative of relatively hospitable lake substrates that would sustain a diverse faunal assemblage.

Most notably, LAA is characterized by high proportions of *C. tricuspis* relative to HAA and MAA. Abundant *C. tricuspis* is associated with water bodies typified by the presence of algal mats comprised of *Spirogyra*, upon which this arcellinid feeds [1, 96, 98, 99]. The presence of *Spirogyra* was not recorded in the lakes comprising this study but was also not specifically sought out during the relatively short time spent with the helicopter at each sample station. Several researchers have interpreted the presence of high percentages of *C. tricuspis* to be a good indication of eutrophic conditions [1, 7, 96]. However, *C. tricuspis* is seasonally planktic [96, 98]

and thus would be expected to drift away from areas where it is often most abundant: along the vegetated margins of a lake. This allochthonous provenance origin hypothesis provides an explanation for the association between high proportions of *C. tricuspis* and relatively low TOC ($\bar{x} = 21.3 \text{ ppm} \pm 10.4 \text{ SD}$) characterizing the samples containing LAA.

Assemblage 4—Diffugiid Assemblage ($n = 4$)

The faunal makeup of the diffugiid assemblage (DA) is dominated by diffugiid species and strains, primarily *D. oblonga* Ehrenberg 1832 strain “lanceolata” ($\bar{x} = 32.7 \% \pm 9.6 \text{ SD}$). Other common diffugiid species and strains includes *D. oblonga* Ehrenberg 1832 strain “oblonga” ($\bar{x} = 9.9 \% \pm 8 \text{ SD}$), *D. protaeiformis* Lamarck 1816 strain “protaeiformis” ($\bar{x} = 4 \% \pm 4 \text{ SD}$), *D. glans* Penard 1902 strain “glans” ($\bar{x} = 5.5 \% \pm 5.7 \text{ SD}$), and *D. elegans* Penard, 1890 ($\bar{x} = 4.3 \% \pm 4 \text{ SD}$). *Cucurbitella tricuspis* (Carter 1856) ($\bar{x} = 13 \% \pm 9 \text{ SD}$) also made up a significant proportion of the fauna. The proportion of centropyxids is notably reduced in this assemblage, with only *C. aculeata* (Ehrenberg 1832) strain “aculeata” ($\bar{x} = 2.5 \% \pm 3 \text{ SD}$) being present in low to moderate proportions. This assemblage is represented by a sample collected from transect W, another from transect E, and two samples from transect S (Fig. 1). The distance between Giant Mine and DA-bearing samples ranges from 3.6 to 19 km ($\bar{x} = 11.7 \text{ km} \pm 7.9 \text{ SD}$). DA occurred at an average sampling depth of 4 m ($\pm 1.8 \text{ SD}$). In contrast to the silty substrates characterizing the first three assemblages, DA-bearing sediments are dominated by a mixture of silt ($\bar{x} = 63 \% \pm 5 \text{ SD}$) and clay ($\bar{x} = 25 \% \pm 11 \text{ SD}$). The range of SDI values for the assemblage (1.88 to 2.27) indicates transitional environmental conditions [10, 87].

The DCA and cluster analysis results show that DA samples are tightly grouped and quite distinct from all other observed assemblages (Figs. 4 and 5). DA-bearing samples are characterized by moderate As concentrations ($\bar{x} = 80 \text{ ppm}$, $\sigma = 72 \text{ ppm}$). However, results of RDA analysis suggest that As is weakly influencing the faunal distribution of the assemblage by showing a negative correlation between the assemblage and As (Fig. 6). The results also revealed a positive correlation between DA and Ba along RDA axis 1 (Fig. 6). Barium is an alkaline-earth metal that is found in over 80 minerals, principally barite (BaSO_4) and witherite (BaCO_3 ; [100]). While barite is considered as an important geochemical proxy of ocean productivity [101, 102], the significance of Ba biogeochemistry in assessing lake history has received little attention [103]. It has been observed though that algal standing crop and Loxodes (ciliated protozoa) has a major influence on Ba biogeochemistry in freshwater (e.g., Esthwaite Water in the United Kingdom; [104, 105]). Therefore, the positive correlation between Ba and DA may be related to lake

productivity, a signal lost in assemblages where the influence of As was very strong.

The faunal makeup of DA samples provides additional evidence of the possible influence of lake productivity on the assemblage. The dominant species in the assemblage, *D. oblonga* “lanceolata,” *D. oblonga* “oblonga,” and *C. tricuspis*, are all characteristic of relatively productive and healthy ecosystems, although the presence of *C. aculeata* strain “aculeata,” *C. constricta* strain “constricta,” and *D. protaeiformis* strain “protaeiformis” indicates a possible low level of environmental stress still exist and thus explains the transitional environmental conditions reflected by the observed SDI for the assemblage.

Assemblage 5—Diffugia Glans Assemblage ($n = 5$)

The Diffugia glans assemblage (DGA) is dominated by *D. glans* Penard 1902 strain “glans” ($\bar{x} = 28.7 \% \pm 14.8 \text{ SD}$), *D. oblonga* Ehrenberg 1832 strain “oblonga” ($\bar{x} = 23.6 \% \pm 15 \text{ SD}$), and *C. tricuspis* (Carter 1856) ($\bar{x} = 11 \% \pm 12 \text{ SD}$). Additional taxa, particularly *C. constricta* (Ehrenberg 1843) strain “aerophila” ($\bar{x} = 10 \% \pm 13 \text{ SD}$), *D. elegans* Penard, 1890 ($\bar{x} = 3.5 \% \pm 4 \text{ SD}$), and *D. protaeiformis* Lamarck 1816 strain “acuminata” ($\bar{x} = 3 \% \pm 5 \text{ SD}$) are also common, but not in all the samples. DGA-bearing samples occurred in lakes from all four transects (Fig. 1). The lakes are located at considerable distances from Giant Mine (17.5 to 23.5 km; $\bar{x} = 20.9 \text{ km} \pm 2.5 \text{ SD}$). Samples associated with DGA were collected from relatively deep sampling depths ($\bar{x} = 6.8 \text{ m} \pm 1.4 \text{ SD}$) and silt-dominated substrates ($\bar{x} = 79.4 \% \pm 16 \text{ SD}$). The SDI values for DGA (1.69 to 1.95) reflect transitional environmental conditions [10, 87].

As with the DA, DGA-bearing samples were closely grouped and plotted distinctly from the other assemblages in the DCA plot (Fig. 5). The RDA analysis results indicate a positive correlation with sampling depth and sedimentary P, and a negative correlation with S1 carbon, S2, carbon, DO, TOC, pH, and Ca (Fig. 6). The correlation between As and DGA is very weak. This result is expected as DGA-bearing samples are characterized by the second-lowest average concentration of As ($\bar{x} = 32.6 \text{ ppm} \pm 27.2 \text{ SD}$) after LAA, although one outlier (BC 2) was characterized by a very high As concentration (905.2 ppm). The correlation of this sample with DGA is most likely due to the fauna of this sample being inexplicably dominated by *D. glans* Penard 1902 strain “glans” (69%). Lakes attributed to DGA are among the furthest from Giant Mine (Fig. 1). This may explain the low As concentrations characterizing DGA samples as the sampled lakes may have been located beyond the impact of the mine’s activities. However, the roughly equitable distribution of arcellinid taxa and moderate SDI values reflect some degree of environmental stress. While the influence of As on DGA is weak, the RDA results show a strong correlation between

sampling depth and DGA samples (Fig. 6). DGA-bearing samples are characterized by the deepest sampling depth among all the assemblage ($\bar{x} = 6.8 \text{ m} \pm 1.4 \text{ SD}$). Therefore, sampling depth may be the variable causing environmental stress in DGA.

Faunas similar to the moderately diverse DGA tend to occur in deep and moderately stressed environments. This assemblage is similar in faunal make-up to the deep water contaminated assemblage (DWCA) reported from Northeastern Ontario [28] and was associated to both deeper waters and contaminated substrates. In the case of DGA, however, the RDA analysis results show DGA samples correlating weakly with As and positively with sampling depth (Fig. 6), thus suggesting that sampling depth, rather than As, is the dominant control over *D. glans* “glans”.

Discussion

The results of cluster analysis and DCA revealed five distinct arcellinid assemblages that were influenced by several environmental parameters, particularly As. The identified faunal assemblages delineated several hydroecologic boundary conditions; between stressed (HAA; SDI values 1.11–2.34), transitional (MAA, DA, and DGA; SDI values 1.65–2.27) and relatively healthy conditions (LAA; SDI values 1.93–2.47).

The results of redundancy analysis (RDA) and partial RDA confirmed that the arcellinid assemblages are influenced by 14 environmental variables, with As being the most statistically significant one, explaining 10.7 % of the total variance in the arcellinid distribution. Measured As levels were notably high in some sampled lake sediments ($\bar{x} = 426.5 \text{ ppm}$; $n = 59$) and even exceeded the ICP-MS method detection limit in one sample (BC 19; >10,000 ppm). Moreover, As concentrations were well beyond the interim sediment quality guideline (ISQG = 5.9 ppm; [61]) in all samples and were over the probable effect level limits (PEL = 17 ppm; [61]) in 91 % of the samples ($n = 54$), with only five samples (BC 11, 35, 45, 52, and 60) having As concentrations below 17 ppm.

In terms of distance from Giant Mine, this study demonstrates that the influence of As on arcellinid distribution as an environmental stressor wanes as the distance between lakes and the mine site increases. Stress-indicating arcellinid species and strains, such as *D. elegans*, *C. constricta* “aerophila,” and *C. constricta* “constricta,” are generally associated with lakes in close proximity to Giant Mine, where the influence of As is most evident. This is particularly clear in the case of HAA, which is generally observed in lakes close to the mine site (average distance from the mine site = 11 km \pm 6.7 SD) and characterized by high As concentrations in their sediments ($\bar{x} = 1090.9 \text{ ppm} \pm 2477.6 \text{ SD}$; $n = 18$). Transitional arcellinid assemblages (e.g., MAA, DA, and DGA) are characteristic of lakes that are located at intermediate distances

from Giant Mine (average distance from the mine site = 15 km \pm 5.6 SD) and characterized by moderate As levels ($\bar{x} = 148 \text{ ppm} \pm 238 \text{ SD}$; $n = 21$). Not surprisingly, the faunal structure of these assemblages is dominated by similar numbers of stress- and health-indicating arcellinid taxa. This may be indicative on a declining influence of As on the arcellinid distribution in these lakes, which is associated with transitional environmental conditions and higher arcellinid diversity in these assemblages. The healthiest arcellinid assemblage (e.g., LAA) characterized lakes that are situated at considerable distances from the mine (average distance from the mine site = 19.3 km \pm 9.5 SD). These lakes are distinguished by relatively low As levels ($\bar{x} = 76.9 \text{ ppm} \pm 123.8 \text{ SD}$; $n = 13$) and diverse arcellinid populations (SDI = 1.9–2.4) that include statistically significant numbers of healthy environment-indicating taxa (e.g., difflugiid species and strains, particularly *D. oblonga* strain “oblonga” and *C. tricuspidis*).

While it is not possible to definitively determine the source of As input in the region by only examining contemporary sediment surface samples, the spatial distributions of high As-level lakes located primarily downwind (north and northwest; ($\bar{x} = 712 \text{ ppm} \pm 1959 \text{ SD}$; $n = 30$)) of the former Giant Mine site provides evidence that these higher As levels are most likely of anthropogenic origin (Fig. 1; [76]). Processing of ore during the early years of the Giant Mine’s operation (1948 to 1958) resulted in very high atmospheric emission of As_2O_3 from the onsite roast stack (thousands of kilograms per day), which subsequently decreased significantly in later years as more stringent environmental laws came into effect [41]. The atmospheric emission of As_2O_3 finally ended with the cessation of the Giant Mine’s activities in 2004. Lakes with relatively lower As concentrations ($\bar{x} = 108.7 \text{ ppm} \pm 203.9 \text{ SD}$; $n = 22$) are mostly located to the east and south of the Giant Mine site (Fig. 1). These results provide strong circumstantial evidence that the high As concentrations from the lakes to the N and NW of the Giant Mine are a direct result of the historical release of As_2O_3 . Elevated As in the underlying bedrock geology of these lakes [106, 107]) are another possible supplementary source. Downcore analysis at the stations with high As levels is required to definitively differentiate between possible anthropogenic (e.g., Giant Mine’s emissions) and naturally derived As in these lakes.

Although As has the most statistically significant control on arcellinid community composition (10.7 %; $p < 0.10$), several other variables, linked to changes in nutrient loading, lake trophic status, and primary productivity, also influence the arcellinid distribution significantly. Partial RDA results confirm that sedimentary P (8.5 %; $p < 0.01$), Ba (6.2 %; $p < 0.01$), and S1 (6 %; $p < 0.01$) are statistically significant controls over the arcellinid distribution and collectively explain (20.7 %) of the total variance. The influence of nutrient loading and lake

primary production are evident in the faunal makeup of DA, DGA and, to some extent, HAA. While the influence of As is particularly strong on the majority of HAA-bearing samples, the RDA plot shows some of HAA-bearing samples correlating closely with S1 carbon along the first RDA axis (Fig. 6). In contemporary lake sediments, S1 carbon corresponds to chlorophyll as well as highly labile geolipids (e.g., fatty acids). High chlorophyll is related to high primary production and lake trophic status [108]. Although S1 carbon values are not particularly high in HAA-bearing samples ($\bar{x} = 46$ mg hydrocarbons/g of sediment ± 1990.4 SD; $n = 29$), the close correlation between S1 HAA in some samples suggest that the lakes corresponding to these samples are slightly more productive and hospitable than the rest of the lakes characterized by HAA. This assessment is corroborated by the presence of significant percentages of *D. oblonga* “oblonga” and *C. tricuspis* in these samples, which thrive in relatively healthy conditions [1]. Similarly, samples correlated with sedimentary P (e.g., DGA) are characterized by high proportions of Arcellinina taxa indicative of a healthy ecosystem. This is expected as P is a limiting macronutrient that is often considered to be the primary cause of eutrophication in lacustrine environments, particularly those impacted by point source pollution (e.g., sewage and/or agriculture [109]). The influence of Ba is most evident in DA, which is dominated by difflugiid species and strains and *C. tricuspis*. An assessment of the significance of Ba biogeochemistry on lake primary production history has received little attention [103]. However, several studies have noted an apparent significant influence of algal standing crop and *Loxodes* (ciliated protozoa) on Ba biogeochemistry in freshwater (e.g., Esthwaite Water in the United Kingdom; [104, 107]). It is therefore concluded that the positive correlation between Ba and DA samples observed here may be related to lake productivity.

Conclusions

The results of this research provide new insight into the sensitivity of Arcellinina to various environmental controls, particularly As, in four transects radiating out from the Giant Mine site, Yellowknife, Northwest Territories, Canada. Of 61 lake surface sediment samples, 59 samples yielded statistically significant arcellininid populations. The distribution of five distinct arcellininid assemblages, identified using Q-mode cluster analysis and DCA, has been quantitatively linked to a number of specific controlling variables using RDA analysis.

Ordinations (RDA) analysis resulted in identification of 14 statistically significant environmental variables, which

collectively explained 57 % of the variance in the arcellininid distribution. Partial RDA analysis provided further confirmation that As had by far the largest influence on the assemblage variance, explaining 10.7 % ($p < 0.01$) of the total variance. This is an important finding that underscores the sensitivity of Arcellinina to As, which was found to be present in many lakes through the region at concentrations significantly above ISQG and PEL guideline levels [61]. The correlation between As and HAA was particularly strong, where As values of up to 10,000 ppm ($\bar{x} = 1090.9$ ppm ± 2477.6 SD; $n = 18$) were observed in conjunction with a *D. protaeiformis* and centropxyid-dominated opportunistic assemblage. As most lakes supporting HAA were downwind (N and W) of the former Giant Mine roaster stack, there is a significant likelihood that the high As in these lakes was industrially derived, a hypothesis that cannot be confirmed without core analysis. Other important factors controlling arcellininid faunal distribution, for which this study has provided new quantitative data, include sedimentary P, which explains 8.5 % ($p < 0.01$) of the total variance, Ba 6.2 % ($p < 0.01$) and S1 6 % ($p < 0.01$).

The demonstrated sensitivity of Arcellinina to environmental As means that they can be used to quantify the impact of this contaminant on lacustrine ecosystem health. Because arcellininids preserve well downcore, the results of this research demonstrate that they can be used to determine baseline lake ecosystem health and associated paleo-As levels, prior to the establishment of the Giant Mine, which is important in the determination of whether the As in these lakes is of anthropogenic or natural origin. For lakes where As is determined to have been of anthropogenic origin, downcore distribution of arcellininid assemblages can also be used to determine whether there has been ecosystem remediation in the decades since the cessation of As emission from the Giant Mine roaster stack.

The preliminary results of this study suggest that Arcellinina hold considerable potential as bioindicators for As contamination in lacustrine ecosystems. The results of this study will be of use to policy makers and planners when evaluating the merit of ongoing and planned remediation programs (e.g., The Giant Mine Remediation Project).

Acknowledgments Funding for this research project was provided by NSERC Strategic Project Grant, NSERC Discovery Grant, and a Department of Aboriginal and Northern Affairs Cumulative Impact Monitoring Program grants awarded to RTP. Additional direct and in-kind funding was provided by the Northwest Territories Geoscience Office and Natural Resources Canada Polar Continental Shelf Program, and the Geological Survey of Canada. We also thank Dr. Graeme Swindles (Leeds, UK) for valuable discussions on the different multivariate ordination methods used in this work. We also extend our thanks to Dr. Paul Gammon for help with the geochemical analyses and interpretations.

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