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ORIGINAL ARTICLE

Tracing groundwater discharge in a High Arctic lake using radon-222

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Abstract In the High Arctic, groundwater fluxes are limited by the presence of continuous permafrost, although it has been hypothesized that there may be localized groundwater flow and hydraulic connectivity beneath large lakes, due to the presence of taliks, or large regions of unfrozen ground. However, due to the logistical difficulty of employing seepage meters and piezometers in deep, ice-covered lakes, relatively little is known about groundwater discharge to polar lakes. One method of assessing groundwater discharge is through the use of geochemical tracers. We conducted a pilot study to quantify groundwater discharge into a High Arctic lake using dissolved radon gas as a geochemical tracer. Lake water was collected in 15 L polyvinyl chloride (PVC) bags with minimal atmospheric interaction from a 25-m deep lake near Shellabear Point, Melville Island, Northwest Territories, Canada. Sample bags were aerated through a closed water loop for 60 min to allow sufficient radon to equilibrate in a coupled air circuit. Radon in air concentrations were measured on a Durridge RAD7 portable alpha spectrometer. The field trial in a remote setting and separate tests with groundwater samples collected from a temperate site demonstrate the utility of the methodology. The limited results suggest that radon levels in the lower water column are elevated above background levels following nival melt in the surrounding watershed. Although these results are insufficient to quantify groundwater discharge, the results suggest subsurface flow may exist, and further study is warranted.

Keywords Groundwater · Arctic lake · Hypersaline · Radon-222 · Permafrost

Introduction

In the Arctic, relatively little is known about the hydrogeological processes of groundwater flow and routing (Woo et al. 2008). Yet, groundwater is an important resource that can influence geotechnical infrastructure such as pipeline and roadway stability, determine the chemical signature of surface water bodies, and impact a range of biological processes including nutrient transport and fishery dynamics (van Everdingen 1990). In most Arctic hydrological budgets, groundwater is assumed negligible due to the presence of continuous permafrost, which restricts flow due to the extremely low hydraulic conductivity of frozen ground (van Everdingen 1990). However, in the High Arctic, lakes and large ponds generate a thermal unconformity at the ground surface due to the high heat capacity of water (Burn 2005). Water above 0°C at the bottom of a lake will prevent lake bottom sediments from freezing, initiating a sub-lacustrine talik that may extend to the base of the permafrost (Burn 2002). Because of this unusual thermal regime, large lakes may receive groundwater discharge, similar to lakes in temperate regions (Winter 1999). If correct, groundwater

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may moderate water-level fluctuations, and supply nutrients and inorganic ions to these lake ecosystems (Hayashi and Rosenberry 2002).

Despite the potential importance of groundwater for Arctic lake systems and water budgets, it is usually not considered due to the inherent difficulty in measuring discharge (Woo 1990). In temperate systems, groundwater flow into lakes is typically measured with seepage meters installed in the bottom sediments (Lee 1977). In Arctic lakes, where ice cover complicates sub-surface moorings, and logistics often prohibit underwater diving, the installation of seepage meters becomes prohibitive. Furthermore, the localized nature of seepage measurements requires a spatial sampling pattern to obtain a representative flux (Burnett et al. 2003). By contrast, the use of geochemical tracers, such as radon, naturally integrates conditions in the lake and effectively eliminates the need for spatial measurements.

Radon (Rn-222) is an inert gas, with a half-life of 3.82 days that is produced during the radioactive decay of radium-226, which has a half-life of 1,622 years. Since radon is inert, and the first gaseous element in the uranium-238 decay series, it will readily diffuse into air instead of remaining bonded in the chemical compound that contained the parent element (Drever 2002). As radium is present in all rock material, it will enter aqueous solution during water–rock interactions. The processes of rock weathering, radioactive decay of radium to radon, and restricted atmospheric interaction, leads to a large concentration of unsupported radon in groundwater (Kraemer and Genereux 1998). Conversely, surface waters typically have low radon concentrations due to the negligible radon content of precipitation. If any radon is present in surface water, it will rapidly diffuse into the atmosphere. This difference allows radon to function as an indirect tracer of groundwater discharge into water bodies and any subsequent physical mixing processes.

With recent advances in detection methods, sampling for radon has become more readily employed in hydrogeological studies (Lee and Kim 2006; Burnett et al. 2001; Gleeson et al. 2009; Schmidt et al. 2009; Burnett and Dulaiova 2003). In an effort to detect the presence of groundwater discharge in a region of continuous permafrost, we conducted a pilot study to test if radon activity could be used as evidence for groundwater discharge into a coastal hypersaline basin in the Canadian High Arctic. While the origin of hypersaline lakes in the Arctic has been debated (Dugan and Lamoureux 2011), one explanation for the accumulation of salt in the lake is the discharge and accumulation of groundwater brines (Ouellet et al. 1989). Although recent work has suggested that the period of tidal influence during isolation of the lake from the ocean is also a viable mechanism for brine accumulation (Dugan and Lamoureux 2011), groundwater discharge was assessed using radon as a tracer to test alternative models. We present a viable field method to conduct radon measurements in remote field settings, and generate an initial dataset to evaluate discharge in this Arctic setting. To our knowledge, this study was the first of its kind in the High Arctic and adds to the sparse limnological literature using radon as a tracer for groundwater discharge (Schmidt and Schubert 2007; Kluge et al. 2007; Schmidt et al. 2009).

Study site

Shellabear Lake (unofficial name, 74°50′N, 113°30′W) is a seasonally isolated marine basin located near Shellabear Point on Melville Island in the Canadian High Arctic (Figs. 1, 2). The lake, which is situated at sea level with a 1 m deep outlet, has an area of 0.59 km^2 and a volume 8.49×10^6 m^3. From November to June, the outlet is obstructed by snow and ice, and the lake is isolated from the ocean. However, open water conditions from June to October permit marine inflow into the lake during the summer. The lake is hypersaline, with a water column salinity of 56 g kg^{-1}.

The lake basin is relatively flat, with a maximum depth of 26 m and a mean depth of 14 m. From approximately June to October, the lake receives water input from the ocean, and from one major river that drains a 17 km^2 watershed. During the winter and until June, the lake is hydrologically isolated from the watershed and atmosphere by a 2 m thick ice cover. In 2009, fluvial inflow began on 16 June, and marine connection did not occur before 1 July (Dugan and Lamoureux 2011).

Geological mapping around Shellabear Lake is limited to regional studies and reconnaissance surficial mapping. The area is underlain by the Devonian Griper Bay formation which is composed of subhorizontal units of sandstone, siltstone and shale. Surficial sediment cover is composed of Quaternary glacial and marine sediments, but the thickness and internal structure at Shellabear Point is unknown (Hodgson et al. 1984).

Methods

In a simple lake system, there are three sources and two sinks of radon. Radon is introduced into the system through groundwater advection, diffusion from the sediments, and in situ production, and is removed by atmospheric loss and in situ decay (Schmidt and Schubert 2007). The objective when utilizing radon as a tracer of groundwater flow is to discern the advection term in the system.

At Shellabear Lake, a field camp was established from 1st to 26th June 2009. To measure radon concentrations,

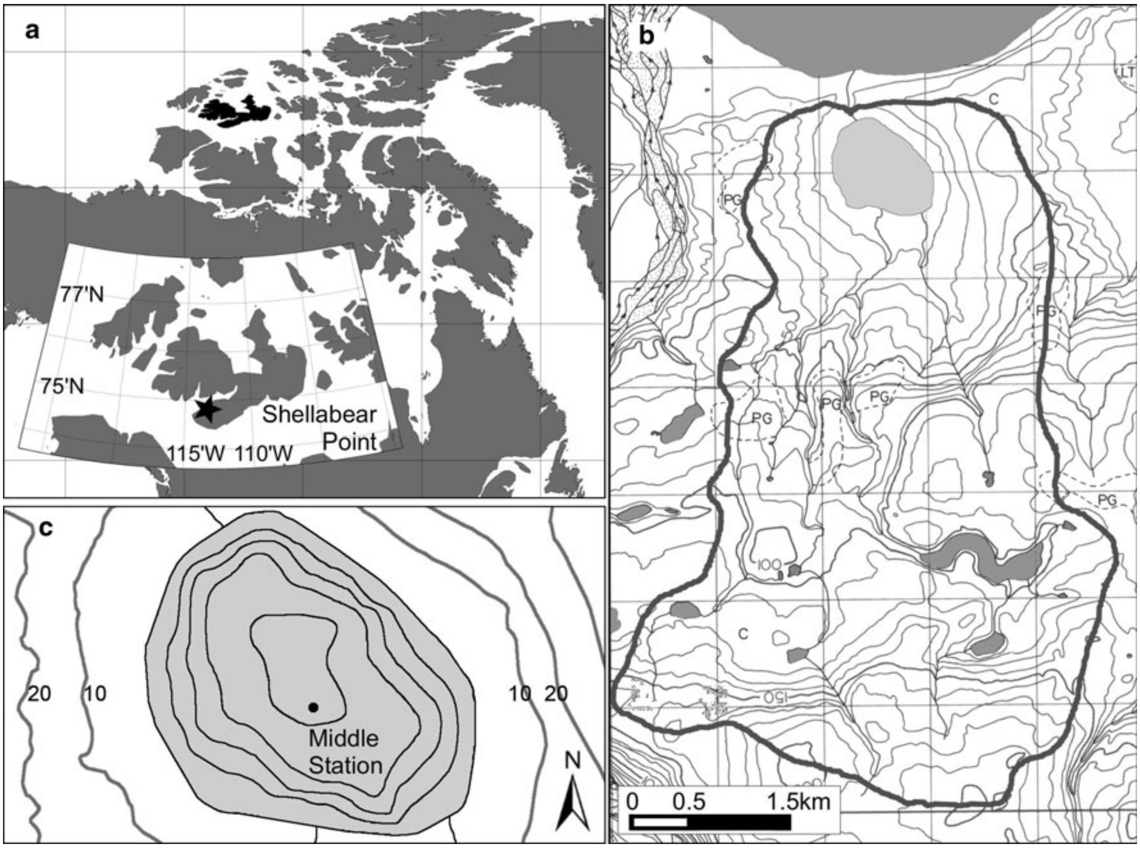

Fig. 1 **a** Location of Shellabear Lake on Melville Island, in the Canadian High Arctic. **b** Shellabear Lake watershed (from NTS map 88 E/15, 1:50,000). Areas of polygons are noted (PG). **c** Bathymetry of Shellabear Lake and location of middle station where sampling took place. Bathymetric isolines and contour lines are at 6 and 10 m intervals, respectively

Fig. 2 A photograph of Shellabear Lake looking north toward Liddon Gulf, Arctic Ocean, taken 13 June 2009

a Durridge RAD7 alpha spectrometer was employed in conjunction with a Durridge RAD AQUA. The intrinsic background of the RAD7 is 0.2 Bq m^{-3}, and the calibration accuracy is 5% (Durridge 2000). The high resolution capability of this detector makes it well suited for a system where low radon concentrations are expected. In addition, the RAD7 was chosen because it allows sample analysis directly in the field. This is a key advantage in a remote environment where samples cannot be returned to the laboratory in less than 1–4 weeks and liquid scintillation analysis is not possible (Zouridakis et al. 2002).

Due to time constraints in the field, the water column was only sampled for radon at a station located at the central, deepest site (Middle Station, Figs. 1, 2). Sampling took place from the ice cover on 4th, 5th, 11th, 18th, 19th, 24th, and 25th June at water column depths of 2, 6, 12, 20, and 26 m. In addition, samples from the river gauging station, approximately 100 m upstream from the lake and the river/lake confluence, were obtained on 24th, 25th and 26th June. Finally, background atmospheric levels were measured on 11th, 23rd and 26th June.

Samples were collected from the lake at depth with a 2-L Kemmerer water sampler. The spigot on the outflow of the Kemmerer was attached to a short section of PVC tubing connected directly to a 15-L PVC bag to avoid atmospheric contamination. The Kemmerer was repeatedly lowered to the required depth, and each sample collected was withdrawn directly to the 15-L bag until the bag was full. The bag was then returned to a nearby shore station which housed the RAD7. The PVC bags were chosen to take advantage of a larger sample size, which improves the minimum detectable radon activity (Stringer and Burnett 2004). To analyse samples, the bag was positioned at height and allowed to drain through the RAD AQUA, which aerated the water. The bottom of the RAD AQUA was equipped with a Micra Plus submersible pump, which returned water back to the 15-L bag, creating a closed loop water circuit. The air intake was positioned at the side of the RAD AQUA, and ran through a desiccant before reaching the RAD 7. The air loop was not purged with radon-free air prior to sampling, as it was decided that background atmospheric levels were sufficiently low that atmospheric radon would not contaminate results. An Onset HOBO external temperature probe was attached to the RAD AQUA to record the temperature of the aerated water. Due to the limited number of available PVC bags, samples were discarded following analysis. Because secondary radon analysis was not preformed, the concentration of radon in the water column supported solely from background radium-226 cannot be distinguished from groundwater supplied radon, or the ‘excess radon’.

Near the end of June, when the river inflow was near peak discharge, samples were analyzed from the lake inlet, and from the river. At this time, no 15 L bags remained intact, so samples were collected in 40 mL vials which allow direct use with the RAD H_2O apparatus. This sample size is intended for higher concentration samples (Durridge 2000).

Each sample was analyzed for at least 1 h, with measurements taken every 10 min to permit adequate time for the sample to equilibrate in the air. Samples were always analyzed within 1.5 h of collection. For each sample, the results from the first 30 min were discarded, and the final readings and uncertainties were averaged for final concentrations in air. A partition coefficient ‘k’ for each sample was calculated from:

$$k = 0.105 + 0.405\mathrm{e}^{(-0.0502T)} \tag{1}$$

where T is the temperature recorded from the external temperature probe. The concentration of radon in water (C_w) was then calculated from the concentration in air (C_air) following (Lee and Kim 2006):

$$C_\mathrm{w} = (C_\mathrm{air} V_\mathrm{air} + k\, C_\mathrm{air} V_\mathrm{w})/V_\mathrm{w} \tag{2}$$

where V_air represents the volume of air in the closed air loop between the RAD H_2O and the sampling bag, and V_w is the volume of water in the sampling bag.

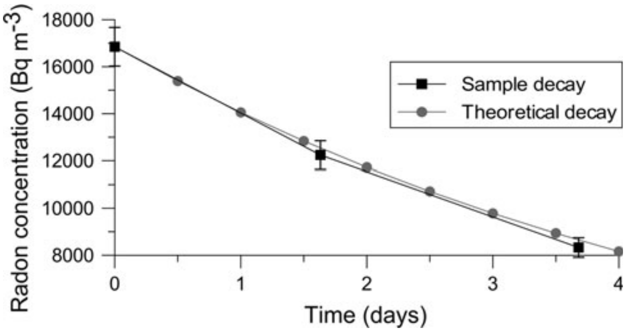

Fig. 3 Radon concentration in groundwater samples allowed to decay over 4 days. The decay curve matches the theoretical decay calculated from a radon-222 half-life of 3.8235 days

To validate the methodology, groundwater samples were collected near Kingston, Ontario. Three, 15 L bags were filled with groundwater from the same well on 28 July 2009. One bag was analysed on each of 28 July, 30 July, and 1 Aug, using identical methods to those performed in the field. The decay of any radioisotope can be modeled with a standard decay equation:

$$C = C_\mathrm{o}\mathrm{e}^{-\lambda t} \tag{3}$$

where C is the new concentration of the isotope, C_o is the concentration at the time of sampling, λ is the decay constant, and t is time in days. The decay constant is calculated based on the species’ half-life. For radon, $\lambda = \ln(2)/3.8235 = 0.181286$. Results show that radon in the bags decayed as predicted by the radon decay curve (Fig. 3), which indicates a lack of atmospheric interaction in the test system. This result confirms that the sampling methodology was sound, and results can be considered accurate.

Without some knowledge of local radon levels in groundwater, it remains impossible to calculate groundwater fluxes. Unlike temperate systems where samples are drawn from nearby wells, continuous permafrost restricts below-ground drilling. This is the foremost challenge in quantifying groundwater flow in Arctic systems. Therefore, the goal of this paper was not to quantify groundwater discharge, but to establish if probable evidence could be demonstrated for its existence.

Results

The concentration of radon in the atmosphere at Shellabear Lake was tested on three separate occasions, and all results reveal atmospheric levels at 0 Bq m^{-3}. Atmospheric background radon levels are likely negligible due to sedimentary bedrock and the lack of local human atmospheric influence. These results permitted analysis of water samples without purging the air lines with radon-free gas beforehand.

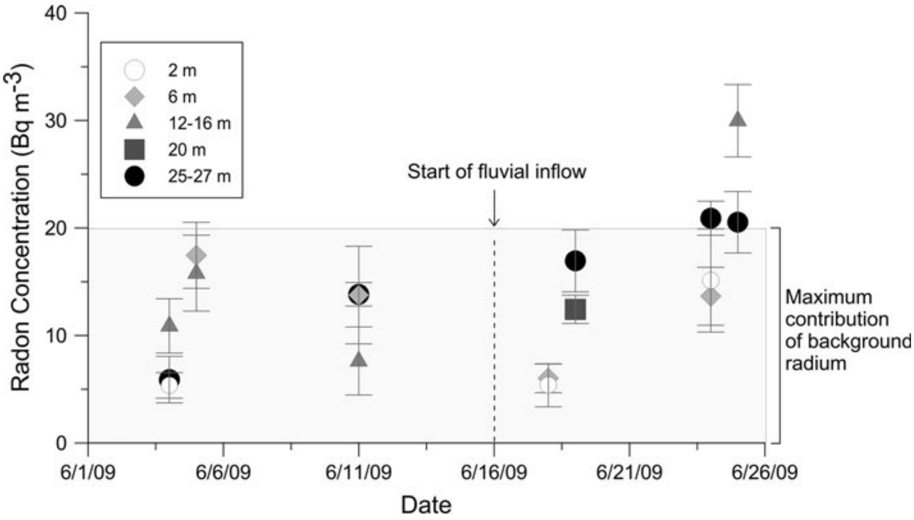

Fig. 4 Radon concentration of water samples from Shellabear Lake. Error bars represent analytical uncertainties associated with the RAD7. The *grey* box represents the hypothesized maximum concentration of background radium in the water column (see text)

Table 1 Radon concentrations of river and lake inlet samples

Date	Location	Concentration (Bq m^{-3})	Uncertainty (Bq m^{-3})
24 June 2009	Lake inlet	65.2	4.7
25 June 2009	Lake inlet	38.0	3.8
25 June 2009	River station	44.4	7.7
26 June 2009	Lake inlet	38.4	5.7
26 June 2009	River station	34.2	4.3

Radon concentrations in the lake were measureable, and higher than background air levels. Samples ranged from 5 to 30 Bq m^{-3}, with the majority of samples falling between 5 and 20 Bq m^{-3}. While a 1–2 m surface ice cover was present for the entirety of sampling, which prevented atmospheric degassing, elevated radon concentrations were not discernable in the epilimnion. Prior to sampling on 24th June, the only discernable depth or temporal pattern was a approximately linear increase in radon concentrations at 25–27 m (Fig. 4). On 24th and 25th June, the mean radon concentration in the water column was elevated as compared to the previous sample dates, largely due to a single high radon concentration at 12 m depth. The radon levels measured in the lake inlet and at the river station were higher than those recorded in the lake. The highest recorded concentration was 65.2 Bq m^{-3} and was obtained at the river outlet on 24th June (Table 1).

Discussion

To the best of our knowledge, using radon as a tracer in the High Arctic has not been attempted prior to this study. This limits any hypotheses that can be derived based on similar environments. However, limnological radon studies have been conducted in temperate locations using similar methodologies (Table 2). From uncorrected data, it appears that the concentration of radon in Shellabear Lake is slightly lower, but near the levels documented in other studies. Although results were not corrected for background radium, the levels are expected to be similar to other lake studies or lower, due to the clastic sedimentary geology of the region (Hodgson et al. 1984). Therefore, we do not expect background radium to be $>\sim$ 20 Bq m^{-3}. Diffusion of radon from the bottom sediments is not considered significant as samples from 25 to 27 m depth were not consistently higher than the remainder of the water column.

The water column in Shellabear Lake is hypersaline below a chemocline at 5 m depth. Due to the high stability of the bottom water, advective mixing by wind is negligible, especially when a surface ice cover is present (9 months of the year). Similarly, mixing from river inflow is minimized due to the strong density contrast between the epilimnion and the hypersaline hypolimnion. Therefore, if a lake-bottom groundwater seep is present, a relatively shallow diffusion profile would be expected. Prior to 24th June, the relatively constant radon concentrations in the water column may signal that radon production is largely supported by background radium. On June 24th, the mean radon concentration in the water column increases. If background radium is assumed to be $\leq$20 Bq m^{-3}, the 16–20 m sample on 25th June is substantially higher than background levels. We consider this rise in radon concentration on 24 and 25th June to be excess radon, which may indicate the presence of groundwater discharge in Shellabear Lake. The excess radon in the upper water column may be associated with the linear increase in radon concentration in the bottom water samples throughout the study period.

Table 2 Comparison between the physical characteristics and radon concentrations of four temperate lakes and Shellabear Lake

Climate	Temperate							Polar
Study site	Schmidt et al. (2010)		Schmidt and Schubert (2007) Lake Waldsee: Germany		Schmidt et al. (2009) Lake Ammelshainer: Germany		Kluge et al. (2007)	This study
	Lake A: Alberta, Canada	Lake B: Alberta, Canada					Lake Willersinn-weiher: Germany	Shellabear Lake: NWT, Canada
Surficial geology	Fine grained sandstone, overlain by glacial drift and fluvial sediments	Shale and silty shale bedrock overlain by glacial till	Moraine of Tertiary lignite, clay, silt and sand layers		Unconsolidated glacial till. Mainly silt, sand and gravel layers		Aquifer consisting of Quaternary sand and gravel	Quaternary glacial and marine sediments underlain by sedimentary bedrock
Area (km^2)	0.11	1.91	0.0034		0.53		0.145	0.59
Vol (m^3)	112,300	1,694,600	7,200		560,000		1,300,000	8,490,000
Z_{max} (m)	2	2.5	4.5		24		20	25
Z_{mean} (m)	1	0.89			10		8	14.4
Max. specific conductivity (μS cm^{-1})	21	31	777	666	780	700	900	80,000
Sampling month	July	July	Oct	Dec	Apr	May	July	June
Radon (Bq m^{-3})	0.5–1.1	11.2–36.6	37.7_E 42.8_H	19.3_E 29.3_H	57.3	57.8	2.6–52.4	5.4–30.0
Ra226 (Bq m^{-3})	Below detection	N/A	19.7_E 17.7_H	6.9_E 14.8_H	18.6	11.1	≤4	N/A
Atmospheric radon (Bq m^{-3})	0.94	9.3	6.6	3.8	5.7	2.8	7.4	0.0

Samples taken from the epilimnion and hypolimnion are denoted by $_E$ and $_H$, respectively

While the highest radon levels were concurrent with the onset of melt and river formation on 16th June, we propose that the river is not the source of excess radon for two reasons. First, the inflowing river water is freshwater, and is restricted to a thin layer between the ice cover and the underlying hypersaline water (Dugan and Lamoureux 2011). Therefore, if the river is supplying excess radon, only the surface water should be enriched. This is contrary to what was observed in Shellabear Lake, as the 2 m sample on 24th June was lower in concentration than the 12 and 27 m samples. Secondly, the overall river discharge was low, reaching a maximum of 0.76 $m^3 s^{-1}$ on 19th June. The quantity of river inflow in proportion to lake water is insufficient to raise radon concentrations of the larger lake volume. However, the difference in sampling methods between lake and river samples must also be taken into account, which limits comparisons, especially at such low overall concentrations.

The increase in radon levels at 25–27 m depth may provide evidence for groundwater input at depth. If there is sub-lacustrine discharge to Shellabear Lake, it is expected to occur following melt, when liquid water is prevalent in the system. Under these conditions, surface water that infiltrates a local groundwater supply could increase the hydraulic gradient and may initiate groundwater discharge. If a talik exists below the lake, water will be directed into the bottom of the lake (Fig. 5). A heuristic rule is that a lake will be underlain by an open talik if the minimum horizontal width of the lake is twice the permafrost thickness in nearby terrain (McGinnis and Jensen 1971). At Winter Harbour, Melville Island (80 km east of Shellabear Lake), temperature measurements obtained from a 3750-m oil exploration well situated 1.2 km inland from the coast indicate a permafrost depth of 557 m (Brown 1972). The width of Shellabear Lake ranges from 800 to 1,000 m and therefore there is a high probability of an open sublacustrine talik. Furthermore, permafrost measurements are based on a 0°C boundary. In an area of saline sediments, freezing point depression due to high salinities can contribute to groundwater flow below 0°C (Pollard et al. 1999).

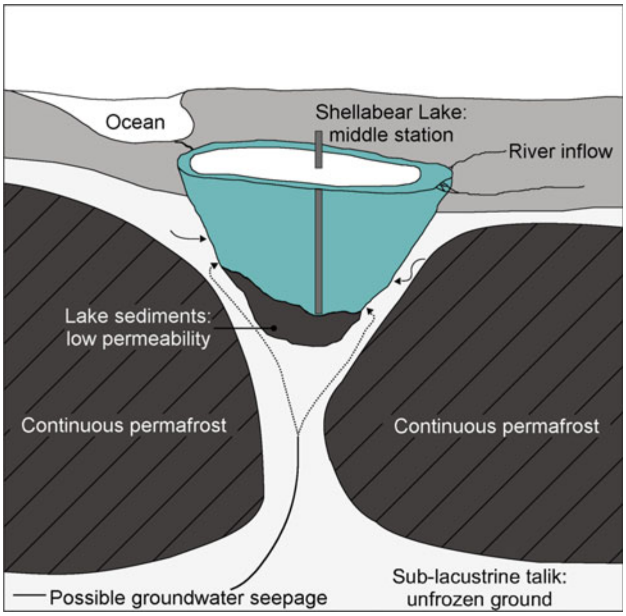

Fig. 5 A hypothesized cross-section of the hydrological characteristics of Shellabear Lake (not to scale). Note the river inflows, the outlet to the ocean, and an open sublacustrine talik. Within the section of unfrozen ground, groundwater discharge may enter the water column at the edges of the basin where low permeability lake sediments are thin or non-existent

At the lake bottom, discharge is often restricted due to the accumulation of thick sediments (Fig. 5). If sediments are predominately silts and clays, the low hydraulic conductivity will restrict inflow (Schmidt et al. 2010). Instead, discharge may enter the lake from the sides, a common feature in temperate lakes (Kluge et al. 2007; Winter 1999).

There are two explanations that may account for the limited range of radon concentrations observed in June 2009. First, sampling was only conducted in the centre of the lake. It may be that discharge enters the lake on the sides, as observed by Kluge et al. (2007), where the radon concentration in the small, shallow basins was relatively high versus sampling undertaken at the deepest location. By the time radon diffuses to the middle of the lake, most excess radon has decayed. Secondly, melt did not begin until mid-June, and the landscape was still partially snow-covered and cold at the end of June. Therefore, it is likely that sampling was conducted too early to discern sustained radon inflow into the lake, and later in the season, when the active layer is thicker and liquid surface water may penetrate into aquifers, more groundwater flow may be expected. However, these explanations are not mutually exclusive and it is clear that the 2009 sampling regime was limited. Expansion of sampling throughout the lake, as well as throughout the melt season will supply more robust results.

Conclusions

The goal of this study was to identify the presence of groundwater flow into a coastal lake in the High Arctic. Shellabear Lake was chosen due to the high brine content, but this methodology can be applied to any lake system. Based on limited data, radon concentrations increased above stable background levels at select depths in the water column after the onset of snowmelt and river runoff. This may denote the presence of sub-lacustrine discharge into the lake. Due to limited sampling, the relative contributions of groundwater discharge, background decay of radium, and radon diffusion from lake sediments cannot be elucidated.

These results raise numerous questions surrounding the presence of groundwater flow in Arctic systems and a more thorough evaluation of radon concentrations in Shellabear Lake and lakes throughout the Arctic is required. Additional sampling near shore and through the melt-season is warranted and would improve the understanding of the origin and flow characteristics of groundwater through continuous permafrost.

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