Noble gas and isotope geochemistry in western Canadian Arctic watersheds: tracing groundwater recharge in permafrost terrain

Nicholas Utting · Bernard Lauriol · Neil Mochnacz · Werner Aeschbach-Hertig · Ian Clark

Abstract In Canada's western Arctic, perennial discharge from permafrost watersheds is the surface manifestation of active groundwater flow systems with features including the occurrence of year-round open water and the formation of icings, yet understanding the mechanisms of groundwater recharge and flow in periglacial environments remains enigmatic. Stable isotopes ($\delta^{18}O$, δD, $\delta^{13}C_{DIC}$), and noble gases have proved useful to study groundwater recharge and flow of groundwater which discharges along rivers in Canada's western Arctic. In these studies of six catchments, groundwater recharge was determined to be a mix of snowmelt and precipitation. All systems investigated show that groundwater has recharged through organic soils with elevated P_{CO2}, which suggests that recharge occurs largely during summer when biological activity is high. Noble gas concentrations show that the recharge temperature was between 0 and 5°C, which when considered in the context of discharge temperatures, suggests that there is no significant imbalance of energy flux into the subsurface. Groundwater circulation times were found to be up to 31years for non-thermal waters using the ^{3}H-^{3}He method.

Keywords Groundwater age · Canada · Stable isotopes · Noble gas · Permafrost

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Introduction

Groundwater circulation and discharge in permafrost regimes represents an enigmatic feature of the hydrological cycle in northern catchments, yet plays an important role in river discharge and maintaining viable spawning and overwintering habitat for sea-run and freshwater fishes (Power et al. 1999; Mochnacz et al. 2010). While groundwater flow is readily documented by the formation of river icings and by reaches of open water throughout the winter, the mechanisms of groundwater recharge, subsurface flow paths and contribution to discharge remain uncertain (Woo and Marsh 2005). The flow of low-salinity perennial groundwater in permafrost terrains is considered to take place in taliks, defined as zones within permafrost regions in which temperatures remain greater than 0 °C. Saline groundwater, for which the freezing temperature is suppressed, can thereby move through permafrost terrain (permafrost being strictly a thermal condition). Accordingly, fresh groundwater can flow perennially within supra-permafrost taliks (above the permafrost table but below the depth of annual freezing), within intra-permafrost taliks (networks or pathways of unfrozen terrain within permafrost) or below the base of the permafrost (Muller 1947; Tolstikhin and Tolstikhin 1976). The distribution of taliks in permafrost basins has been well studied (French 1996; Carey and Woo 2005) and clearly linked to local climate, insolation and the conduction of heat into the soil and rock by thermal diffusion. However, the advective flow of heat into the subsurface through groundwater recharge may play a role in the formation and stability of taliks.

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Groundwater flow in regions of continuous permafrost commonly occurs in karst carbonate and evaporate bedrock, with recharge occurring by diffuse flow or through dolines (Ford and Williams 2007). Karstic groundwater systems are known in numerous areas of northwestern Canada such as along the Firth River in northern Yukon (Clark and Lauriol 1997), rivers in the Norman Wells region of the Northwest Territories (NWT; Michel 1986), the Nahanni River (Brook and Ford 1980), and near Great Bear Lake, NWT (Van Everdingen 1981). In northeastern Canada, groundwater drainage occurs in permafrost on carbonate Akpatok Island (Lauriol and Gray 1990). In Alaska, there are groundwater-fed streams on the northern coast near Prudhoe Bay (Craig and McCart 1975). Many of these systems were established when the climate was warmer prior to glaciation, yet persist under the colder present conditions (Ford and Williams 2007). In the Canadian High Arctic, evidence for groundwater flow is restricted to hypersaline settings. Springs on Axel Heiberg Island (McKay et al. 2005; Omelon et al. 2006 and Pollard 2005) discharge a high total dissolved solids (TDS) mixture of subglacial water and water from a talik below a lake. These waters gain their salinity by flow through dissolution porosity in evaporite rocks. Grasby et al. (2003) describe saline perennial springs discharging supraglacially on Ellesmere Island. Groundwater may also flow through sands and gravels where conditions are appropriate, for example pro-glacial groundwater discharge in Greenland (Scholz and Baumann 1997).

The occurrence of river icings (*aufeis* in German, *naledi* in Russian or *glaçage* in French) is evidence for perennial groundwater discharge in permafrost regions. Icings occur when river ice freezes to the base, blocking discharge and building pressure such that the ice is breached and surface pooling occurs (Carey 1970). They were first described in Russia (Wrangel 1841; Anisimova et al. 1973), and have since been described in many polar areas such as the Brooks Range, Alaska (Yoshikawa et al. 2007); pro-glacially in Svalbard (Bennett et al. 1998); in Finland (McFadden 1990); and in Iceland (Venzke 1988)—see electronic supplementary material (Fig. 1, ESM) for map. In Canada, icings have also been observed in numerous areas. The largest icing in Canada forms annually along the Firth River, Yukon, where 12 % of basin groundwater discharge from the carbonate catchments in the British Mountains is retained annually (Clark and Lauriol 1997). In central Yukon, icings form in the North Fork Pass area (Hu and Pollard 1997). In the Canadian High Arctic, icings form at the Axel Heiberg Island hypersaline springs (Pollard 2005) and in proglacial settings such as the Akshayuk Pass of southern Baffin Island (Lacelle et al. 2006).

In smaller mountain streams (width <20 m) perennial groundwater sources maintain ice-free or below-ice discharge (Prowse et al. 2006). These areas provide spawning and overwintering habitat for northern fishes such as sea-run Dolly Varden (*Salvelinus malma*; Power et al. 1999; Mochnacz et al. 2010). The understanding of groundwater flow in permafrost terrain is also of importance to mining development for determining dewatering needs as well as assessing potential environmental impacts (Stotler et al. 2009).

The study of groundwater in permafrost is challenging given the limited infrastructure and very short field seasons. These conditions favour samples from baseflow discharge and perennial groundwater springs, combined with the use of geochemical and isotope tracers to elucidate recharge conditions and flow paths. Noble gas and isotopic techniques are well suited to studies of the periglacial environment given the effects of temperature on their fractionation and partitioning (e.g., Aeschback-Hertig et al. 2000). Stable isotopes ^{18}O and D of water in periglacial watersheds can record seasonal variations as well as recharge elevation and paleo-effects (Clark et al. 2000, 2001; Michel 1986). A groundwater body with a depleted signature of ^{18}O and D relative to surface water has been interpreted to be the result of recharge at higher elevation or as a result of recharge being dominated by snowmelt (Clark et al. 2001; Michel 1986). Alternatively, ^{18}O has been used as a tracer of different water sources (Hayashi et al. 2004).

The concentrations and isotopic ratios of dissolved inorganic carbon (DIC) can be used to determine recharge conditions. In most cases during recharge, water dissolves CO_2 from the soil where it is produced from the decomposition of organic matter. Vegetation in Yukon and NWT is primarily of the C_3 variety. Vegetation of the C_3 variety uses the RuBisCO enzyme to catalyse CO_2 respiration, which results in vegetation with a $\delta^{13}C$ of between −24 and −30 ‰ (Vogel 1993). When CO_2 produced by the decomposition of C_3 vegetation dissolves in water, the $\delta^{13}C_{DIC}$ is typically ca. −27 ‰ (Clark and Fritz 1997). The acidity in the water is then consumed during weathering of carbonates that have a $\delta^{13}C$ of 0 ‰, which enriches the DIC. Open system conditions exist where there is a continual supply of CO_2 so that there is equilibrium between soil CO_2 and DIC, for example in shallow soils with abundant carbonate and organic material. In this case, the $\delta^{13}C_{DIC}$ is controlled by the pH. Under open system conditions, the final $\delta^{13}C$ is generally between −15 and −18 ‰. Under closed system conditions water dissolves CO_2 during recharge and then weathers carbonate minerals along its flow path; the end $\delta^{13}C_{DIC}$ will be close to −12 ‰. Greater enrichments can be generated through continued exchange between DIC and the carbonate matrix, and through incongruent dissolution of dolomite. Due to the greater partial pressure of CO_2 in soil, groundwater recharge through soils dissolves more CO_2 and so carries a greater weathering potential than it would under atmospheric air conditions. Based on these reactions that occur during recharge, the $\delta^{13}C$ of DIC combined with pH and major ion geochemistry are useful tracers of groundwater recharge (Clark and Fritz1997).

Carbon, oxygen and hydrogen isotopes can indicate environments of groundwater recharge, while noble gases can inform on timing and temperature of recharge. During recharge, the gas composition of groundwater is determined by the atmospheric composition and the salinity

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and the temperature of the water. Once below the water table, dissolved gases do not exchange with the atmosphere and the noble gas concentration is preserved (Kipfer et al. 2002). The differing solubilities of noble gas with recharge temperature have been used in temperate regions to determine mean annual temperature in the recharge environment and to reconstruct past climates (Aeschbach-Hertig and Solomon 2012). In permafrost catchments, the noble gas record of recharge temperature can potentially provide constraints on the seasonality of recharge. In permafrost regions where the mean annual air temperature is below 0 °C, the groundwater recharge temperature can be compared with the temperature at discharge to determine if recharging groundwater advects heat into the subsurface.

Noble gas isotopes produced by radioactive decay, including ^{3}He from tritium, ^{4}He from alpha-decay and ^{40}Ar from ^{40}K provide non-reactive tracers of groundwater circulation rates. The short half-life of tritium decay (12.3 years) is useful for groundwaters actively circulating in permafrost, providing corrections for geogenic ^{3}He production can be made (Lucas and Unterweger 2000).

Groundwater in permafrost watersheds of the northwestern Canadian Arctic

This article compiles results from two previous publications and a PhD thesis that presented data on groundwater from western Northwest Territories and Yukon Territory, Canada. Groundwater discharge has been studied in the Central Mackenzie Valley, NWT (White Sand Creek, Hodgson Creek, Smith Creek, Canyon Creek, Gibson Creek, Gayna River); and along the Fishing Branch River, Yukon and Firth River, Yukon (Fig. 1). Results from the Central Mackenzie Valley are from Utting (2012), a PhD thesis, while results from Fishing Branch River are from Utting et al. (2012) and those from Firth River are from Clark and Lauriol (1997). The study areas are located over a wide geographic area from 63° N to 68° N and 123 °W to 139 °W.

In the areas studied in this publication, the average annual air temperature ranges from −3.2 °C in Fort Simpson to −9.0 °C in Old Crow, allowing permafrost to persist. Total annual precipitation ranges from 265.6 mm at Old Crow to 369 mm at Fort Simpson (Environment Canada 2010). The southern part of the study area is located in extensive discontinuous permafrost which progressively becomes continuous permafrost in the northern part of the study area (NRCan 2003; Fig. 1). Permafrost thickness ranges from <10 to 100 m (Smith and Burgess 2002).

The groundwater systems included in this study are located in the Interior Platform or the Cordilleran orogen geologic provinces of Canada (NRCan 2004). Groundwater flow occurs mostly in Devonian carbonate and evaporite bedrock. Dissolution has formed karst allowing for significant groundwater flow (van Everdingen 1981; Hamilton and Ford 2002; Cinq-Mars and Lauriol 1985). Karstification processes have been occurring since at least the Cretaceous

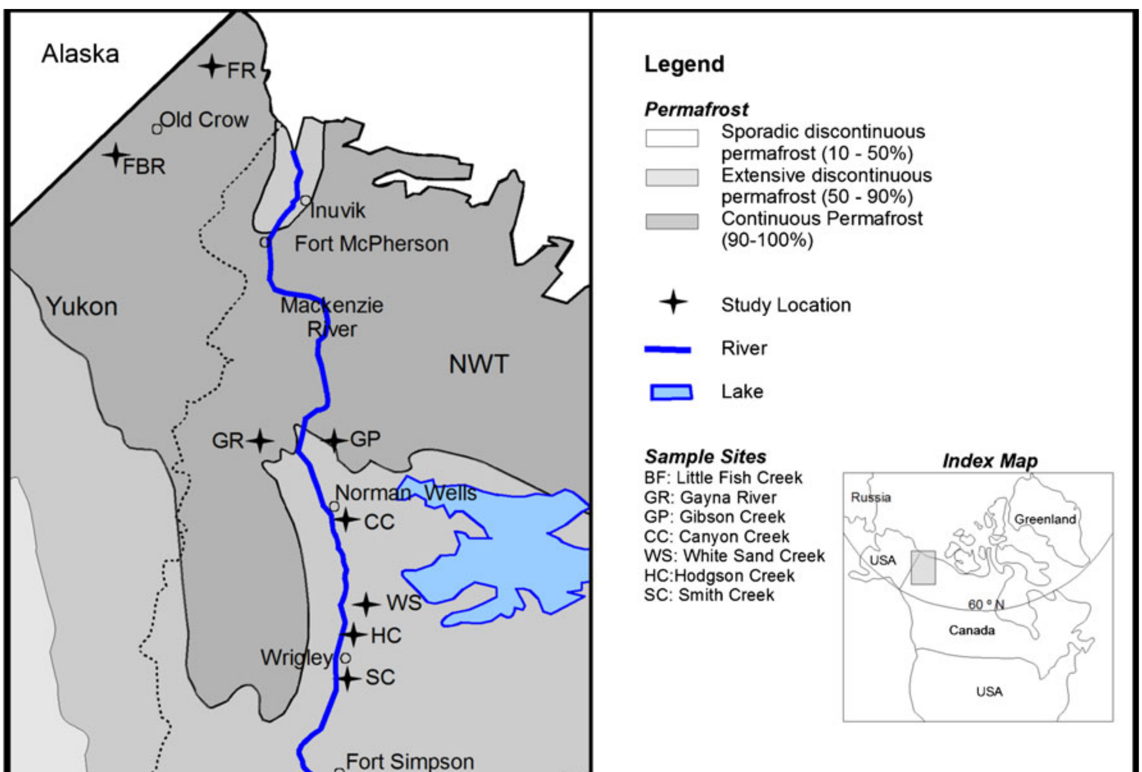

Fig. 1 Distribution of permafrost in the Yukon and the Northwest Territories of Canada, and sampling sites (Permafrost distribution from NRCan 2003)

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but may have slowed during the Pleistocene glaciations (van Everdingen 1981). The groundwater systems included in this study, which are located in the Northwest Territories, were glaciated during the Pleistocene, while the sites in Yukon were not glaciated (Duk-Rodkin 1999).

Field work and analytical methods

Several of the groundwater systems presented in this study were sampled during the past two decades from locations with known perennial groundwater discharge (Mutch and McCart 1974; Michel 1977, 1986). Groundwater samples were obtained from springs while additional surface water samples were collected. More recent sampling was conducted along the Firth River in June 1993 and June 1994 and along the Fishing Branch River in June 2006, July 2007 and August 2008. In August and September of 2007, some ^{18}O and D samples were collected from streams in the Central Mackenzie Valley (Gayna River, Gibson, Hodgson, White Sand and Smith creeks). In March 2008, groundwater and surface water samples were collected in the Central Mackenzie Valley, with further surface water samples being collected in June 2008 for ^{18}O and D. Along Smith Creek perennial groundwater springs were sampled with water temperature of up to 13.8 °C; this maintains open water all year in the creek near the springs. Similarly, along Gibson Creek warm groundwater (8.2 °C) results in open water which flows into the creek maintaining flow in winter. Along White Sand Creek, Hodgson Creek, Gayna River and Canyon Creek groundwater springs were sampled along the banks. Along Smith Creek, Gibson Creek, White Sand Creek, Hodgson Creek, Gayna River and Canyon Creek, the groundwater springs were sampled for major ions, $^{13}C_{DIC}$ and ^{18}O and D; springs from Smith Creek, White Sand Creek and Gayna River were sampled for noble gases and tritium. Surface waters collected along these watercourses were analysed for ^{18}O and D. Details on sample collection and analysis can be found in Clark and Lauriol (1997) for Firth River; Utting et al. (2012) for Fishing Branch River; and Utting (2012) for Central Mackenzie Valley.

As these groundwaters have temperatures above the mean annual air temperature, they could all be classified as thermal waters (White 1957). However, this type of definition is not very useful as most of these groundwaters do not have deep flow pathways where they gain heat in the subsurface. Any separation of thermal vs. non-thermal is somewhat arbitrary, but can be useful for the sake of discussion. In this manuscript, the groundwaters from Smith Creek and Gibson Creek are classified as being thermal, because of their higher temperature and higher dissolved ion concentrations.

Noble gases

Rapid exchange of noble gases with the atmosphere constrains the sites and methods of sampling. In the absence of piezometers and observation well networks in these remote settings, groundwater springs represent the best sites for noble gas sampling, using both passive diffusion gas samplers (Sanford et al. 1996; Manning et al. 2003) and water samples in annealed copper tubes. Details on the sampling and laboratory methods can be found in Utting et al. (2012) and Utting (2012). At each site sampled for noble gases, 500 ml of water was collected for tritium analysis by decay counting after electrolytic enrichment. This analysis was conducted at the University of Waterloo, Canada.

Noble gas samples were analysed at Heidelberg University, Germany, and the University of Ottawa, Canada. At Heidelberg University, samples were analysed for all noble gases using a GV 5400 mass spectrometer, following procedures based on Friedrich (2007). At the University of Ottawa, analysis was conducted on two noble gas lines. Analyses of helium isotopes and neon concentrations were conducted on a MAP 215–50 noble gas mass spectrometer using standard noble gas extraction and analysis techniques (Mohapatra and Murty 2000). Analyses for argon and krypton were conducted using an isotope dilution noble gas line with a quadrupole mass spectrometer based on the design of Poole et al. (1997).

Results and interpretation

Groundwater geochemistry

As presented in the preceding, groundwater circulation within permafrost appears to occur preferentially through secondary porosity associated with evaporite or carbonate karst terrains. First order insights to groundwater circulation can then be derived from major ion geochemistry, which provides a basis for subsequent investigations of isotopes and noble gases. Groundwaters tend to have higher dissolved solids than overland runoff derived surface water due to greater water-rock reaction time, enhanced by carbonic acid derived during transit through soils in the recharge area. The dissolved ions in groundwater may also reflect the groundwater flow path. The carbonate component of groundwater reflects the recharge conditions where CO_2 is dissolved and subsequently weathers mineral material.

Water geochemistry measurements for the various study areas are presented in Table 1 and shown in Fig. 2. Geothermal groundwaters from Smith Creek and Gibson Creek catchments have higher dissolved solids and are dominated by calcium, sodium, chloride and sulphate ions. The high proportion of sodium and chloride in Smith Creek groundwater is attributed to the dissolution of halite (Michel 1986). The other groundwater systems are non-thermal and are dominated by calcium, sulphate and bicarbonate. These ions originate from the weathering of calcite and gypsum bedrock. The higher sulphate relative to bicarbonate in some of the non-thermal groundwaters is presumably due to greater interaction with gypsum rich bedrock in the subsurface.

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Table 1 Concentration of major ions, $\delta^{18}O$, δD, $\delta^{13}C_{DIC}$, $\log(P_{CO2})$ and calcite saturation index. *Smith Creek*, *White Sand Creek*, *Hodgson Creek*, *Canyon Creek* and *Gayna River* results from Utting (2012), *Fishing Branch River* results are from Utting et al. (2012) and *Firth River* results are from Clark and Lauriol (1997)

Creek and sample ID[a]	Latitude (°N)	Longitude (°W)	pH	T (°C)	Ca (Concentrations in mg/l)	K	Mg	Na	Cl^-	SO_4^{2-}	HCO_3^{1-}	$\delta^{18}O$ (VSMOW)	δ^2H	$\delta^{13}C_{DIC}$ (VPBD)	log (P_{CO2})	$SI_{Calcite}$	DOC (ppm)	$\delta^{13}C_{DOC}$ (VPBD)
Smith Creek																		
2008-SCW-3.1-HS-P	63.180	123.335	7.3	13.8	261	7.1	81.7	400	560	790[c]	238	−23.0	−179.0	−7.8	−2.0	0.12	1.5	−26.1
2008-SCW-3.2-HS-P	63.180	123.336	7.5	7.9	324	8.3	99.4	472	651	1,008[c]	237	−23.1	−178.5	−6.8	−2.3	0.37	1.2	−26.5
2008-SCW-4.1-HS	63.181	123.336	7.1	10.3	226	5.8	68.4	283	348	667[c]	258	−23.2	−181.5	−8.1	−1.9	−0.03	1.2	−26.1
2008-SCW-4.2-HS	63.181	123.336	7.4	5.5	157	3.8	50.2	150	189	380	273	−21.6	−171.6	−8.8	−2.1	0.05	11.0	−26.9
White Sand Creek																		
2008-WSW-3-S	63.619	123.607	7.6	3.8	47	0.9	25.1	13.9	7	53	216	−22.4	−174.3	−8.4	−2.4	−0.16	2.1	−26.2
Hodgson Creek																		
2008-HCW-2-S	63.334	123.442	7.5	2.2	67	1.0	28.0	4.4	2.1	71	249	−23.1	−181.8	−9.9	−2.3	−0.07	1.6	−25.8
Canyon Creek																		
2008-CCW-1-S	65.220	126.528	7.4	0.5	140	1.5	51.1	9.3	6.2	348	249	−22.0	−176.8	−7.4	−2.2	−0.04	2.9	−26.1
Gibson Creek																		
2008-GPW-5-HS-P	65.708	127.886	7.3	8.2	316	2.1	102.5	15.5	13.8	1,127	184	−24.2	−188.5	−7.7	−2.2	0.08	1.7	−26.3
Gayna River																		
2008-GRW-1-S	65.297	129.357	7.5	3.1	72	0.6	22.6	11.9	1.1	90	235	−22.3	−174.5	−12.8	−2.2	−0.13	4.3	−27.3
Fishing Branch River																		
2006 Livingstone[b]	66.488	139.323	7.7	4.2	43.8	bld	9.9	4.4	4.4	10.8	182	−22.5	−174.7	−11.31	−2.6	−0.13	2.8	−26.4
2006S4	66.501	139.353	7.6	2.3	47.4	0.3	9.4	2.9	1.7	12.0	186	−23.2	−187.2	−10.81	−2.5	−0.22	2.6	−26.1
2006 BC	66.500	139.371	7.8	4.7	47.2	0.3	10.2	3.9	2.4	18.3	122	−21.3	−163.6	−7.36	−2.8	−0.16	7.5	−26.3
2006 CS[b]	66.527	139.305	7.8	5.7	39.3	1.3	10.4	5.9	5.5	10.9	180	−22.3	−170.9	−9.79	−2.7	−0.06	3.2	−26.2
2007 Upper[b]	66.518	139.371	7.9	4.3	45.7	0.3	9.3	2.6	2.6	20.7	187	−21.9	−168.8	−10.13	−2.7	0.09	1.6	−26.3
2007 BC	66.500	139.371	7.9	5.6	45.1	0.3	10.1	3.4	3.4	24.7	176	−21.5	−166.4	−9.72	−2.8	0.08	1.6	−26.6
2007 Redd	66.508	139.369	8.1	7.2	45.8	0.3	10.3	3.4	3.3	25.1	144	−21.5	−166.4	−9.43	−3.0	0.22	1.7	−25.9
2007 Gossage	66.512	139.379	7.9	7.8	46.4	0.3	10.5	3.4	3.4	24.7	139	−21.5	−168.0	−9.75	−2.9	0.03	1.3	−26.5
2007 Livingstone	66.488	139.323	7.7	4.3	43.1	0.4	10.6	4.1	5.5	15.9	144	−21.9	−170.6	−9.17	−2.7	−0.24	1.4	−26.3
2007Ca	66.527	139.305	7.4	6.1	41.2	0.4	11.2	5.3	7.3	15.3	174	−21.8	−168.2	−8.75	−2.3	−0.45	1.6	−26.5
2008 Upper[b]	66.518	139.371	8.4	4.2	49.7	0.3	10.0	2.8	2.6	22.5	144	−21.7	−167.7	nm	−3.1	0.78	nm	nm
2008 BC	66.500	139.371	8.3	4.9	50.3	0.3	11.2	3.8	3.3	26.8	135	−21.6	−167.0	nm	−3.3	0.39	nm	nm
2008 CS[b]	66.527	139.306	nm	5.5	44.6	0.3	11.8	5.7	7.3	17.2	145	−21.8	−169.6	nm		nm	nm	nm
Firth River																		
SS 2SP	nm	nm	7.4	0.0	29.7	0.3	9.5	0.2	0.6	0.0	118	−20.2	−154.3	−5.92	−2.4	−0.90	nm	nm
LuC 1	nm	nm	7.5	3.0	44.1	0.0	5.5	0.2	1.0	9.3	144	−22.9	−174.6	−5.21	−2.4	−0.58	nm	nm
MB-3 SP-2	nm	nm	7.5	3.5	64.2	0.0	3.1	0.1	nm	17.3	178	−22.4	−176.3	−8.28	−2.3	−0.30	nm	nm
TrC 1sp	nm	nm	7.6	2.0	44.2	0.5	5.2	0.2	0.8	21.1	124	−22.1	−171.6	−3.3	−2.6	−0.54	nm	nm
TA ISP-3	nm	nm	7.6	5.0	48.4	0.8	2.82	0.17	nm	3.9	nm	nm	nm	−8.11	−2.5	−0.29	nm	nm
MB-1 SP-1	nm	nm	7.8	3.0	62.6	0.1	3.1	0.1	nm	17.8	172	−22.4	−176.4	nm	−2.7	−0.01	nm	nm
TrC 2sp	nm	nm	7.9	0.0	39.5	0.1	2.9	0.0	0.6	0.0	118	−21.6	−163.3	−3.79	−2.9	−0.31	nm	nm
MB-5 SP-1	nm	nm	7.9	3.5	75.5	0.0	3.6	0.2	nm	29.6	210	−22.0	−174.4	–	−2.7	0.22	nm	nm
FiT 1	nm	nm	7.2	0.5	113	0.7	9.1	0.2	1.0	185.0	160	−22.8	−175.0	−5.56	−2.1	−0.59	nm	nm
FiT 2	nm	nm	7.7	3.0	107	0.6	8.1	0.1	0.9	160.1	149	−22.4	−171.9	−4.57	−2.6	−0.11	nm	nm

[a] Creek and sample ID: Where available year of sampling is given preceding the sample location name in the sample ID

[b] Indicates average of multiple values

[c] Indicates SO_4^{2-} concentration determined from elemental sulphur concentration

nm not measured. *bdl* below detection limit

Oxygen-18 and deuterium

Figure 3 shows δD and $\delta^{18}O$ values measured from groundwater and surface water along the eight watercourses. Samples from the Central Mackenzie Valley were compared to the meteoric water line from Fort Simpson ($\delta D = 7.6\delta^{18}O - 2$ ‰; Hayashi et al. 2004). Samples from the Fishing Branch River and Firth River were compared to the meteoric water line from Old Crow ($\delta D = 6.8\delta^{18}O - 21.5$ ‰, Utting et al. 2012).

Discharging groundwater is expected to have a nearly constant δD and $\delta^{18}O$ composition throughout the year, whereas the composition of surface water varies seasonally. In general, groundwaters along Smith Creek, Gibson Creek, Hodgson Creek, Canyon Creek, White Sand Creek and Gayna River have lower δD and $\delta^{18}O$ values than surface water collected in these reaches (Fig. 3). Surface waters along these creeks were collected in March and June. The composition of surface water changes through the year but remains enriched compared to groundwater. Groundwater samples from the Fishing Branch River and Firth River watersheds have similar isotopic composition to the surface water. Groundwater in the Fishing Branch has a similar isotopic composition to average precipitation (Utting et al. 2012). The similar isotopic composition of surface water to groundwater is due to the very high contribution of groundwater to the river.

Groundwater recharge: DIC, calcite saturation and $\delta^{13}C$

Groundwater recharged through soils gains considerable weathering potential from the dissolution of CO_2, which is subsequently attenuated through carbonate dissolution. Groundwater samples are near calcite saturation with indices (log IAP/K_{cal}) of −0.90 to 0.78. In Fig. 4, the DIC and pH of groundwater samples are shown along with the saturation curves of calcite and dolomite and the open and closed system weathering trajectories for initial soil partial pressure of CO_2 (P_{CO2}) values of $10^{-2.5}$ and $10^{-3.5}$ atm. Higher P_{CO2} values are more commonly associated with open system weathering reflecting the continual supply of CO_2 from decomposing organic matter in the soil. The lower P_{CO2} of closed system conditions reflects the reduction of acidity by weathering. Given that groundwater recharge is dominantly through soils, they will, under most circumstances, have higher P_{CO2} values than surface waters. The $\delta^{13}C$ of DIC in groundwater can be used as a complementary tracer. In general, a $\delta^{13}C_{DIC}$ near −17 ‰ suggests open system recharge through organic soil. Higher values can be associated with closed system weathering (below the water table) and/or incongruent dissolution of dolomite along the flow path, or possibly a result of methanogenesis in the subsurface (Clark and Fritz 1997).

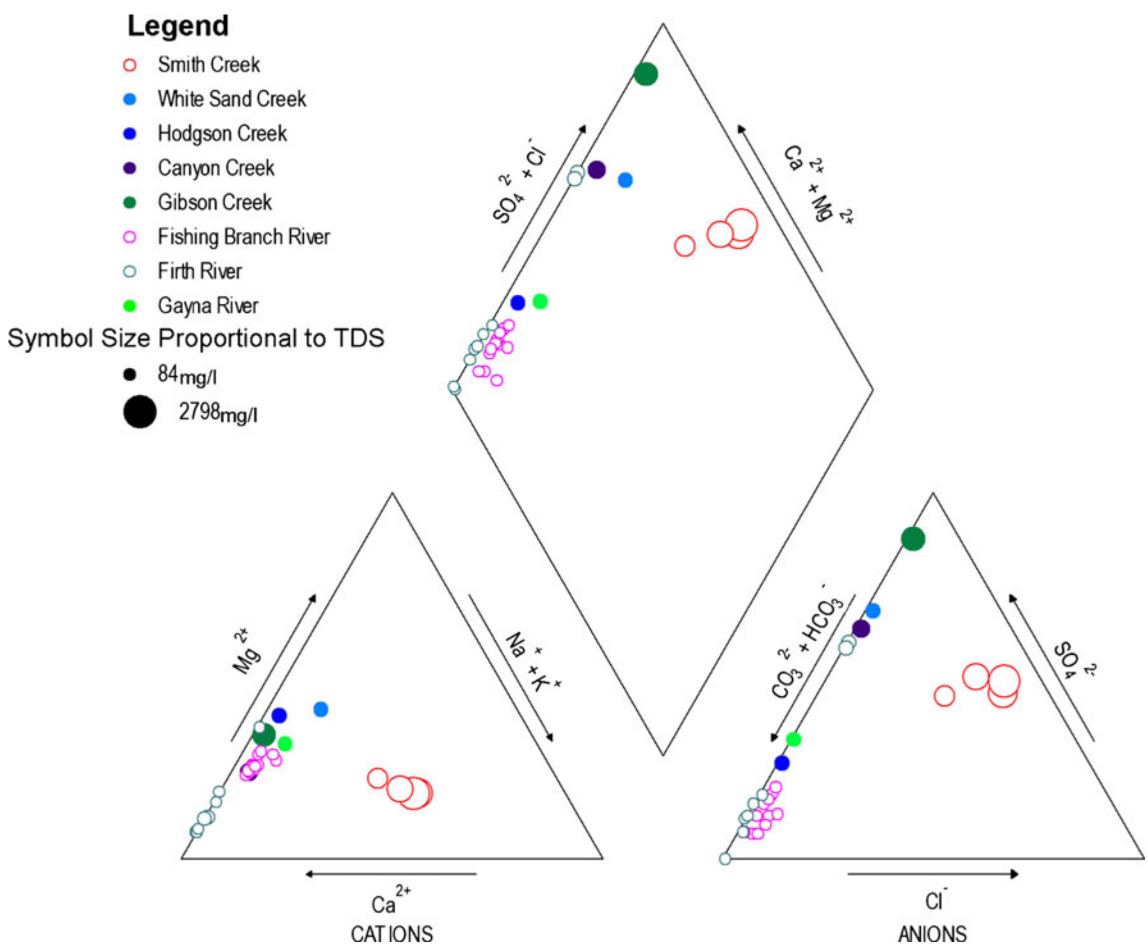

Fig. 2 Piper plot of major ion chemistry from groundwater samples

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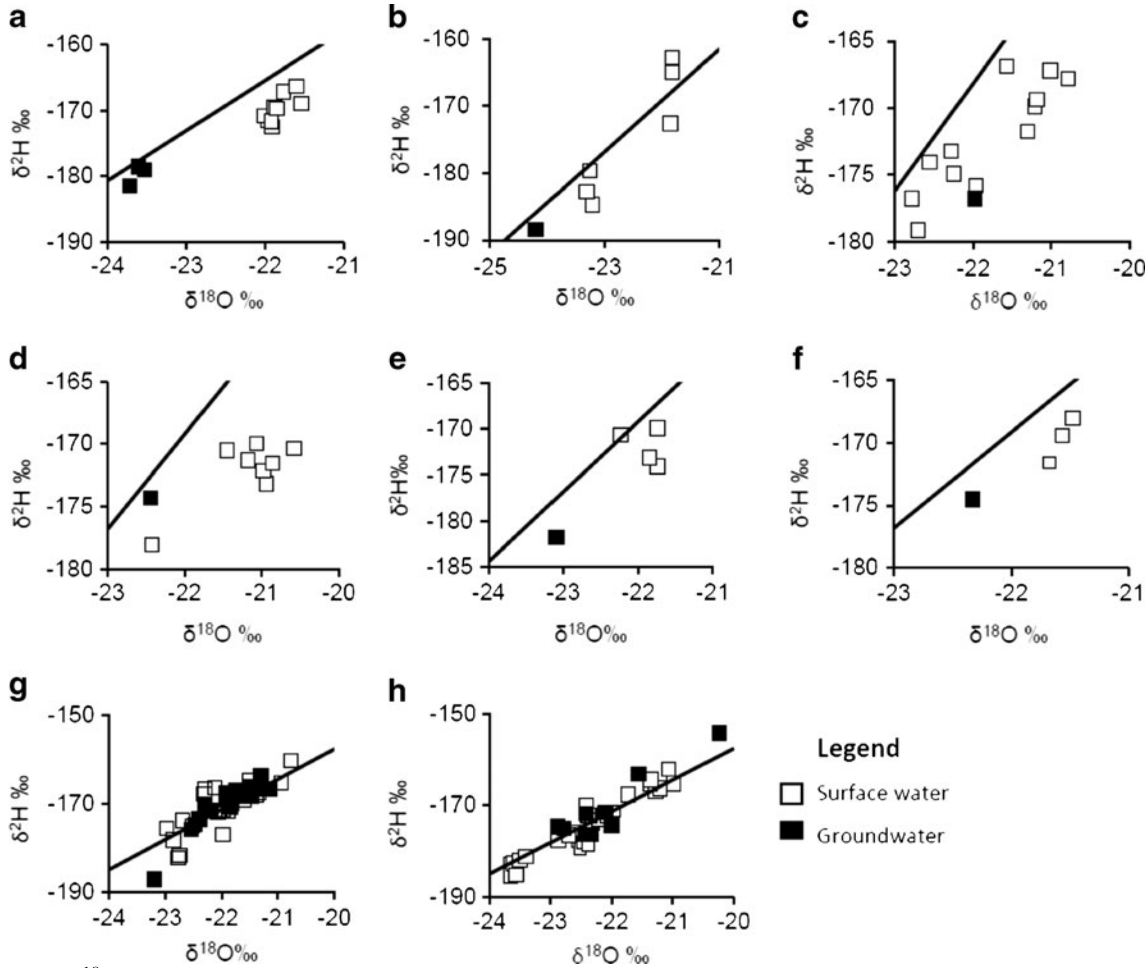

Fig. 3 δ D vs. δ^{18}O isotope results for water samples **a** Smith Creek, **b** Gibson Creek, **c** Canyon Creek, **d** White Sand Creek, **e** Hodgson Creek, **f** Gayna Creek, **g** Fishing Branch River, **h** Firth River. Plots **a**–**f** used the local meteoric water line for Fort Simpson, (δD=7.6δ^{18}O - 2, Hayashi et al. 2004); Plots **g** and **h** used the local meteoric water line for Old Crow (δD=6.8δ^{18}O – 21.6; Utting et al. 2012)

Groundwater samples have an average P_{CO2} of $10^{-2.3}$ atm, which is consistent with recharge through organic soil. The $\delta^{13}C_{DIC}$ of groundwater ranges from –3.3 to –12.8 ‰ (see Table 1). The more depleted $\delta^{13}C_{DIC}$ values are consistent with closed system weathering of carbonate rocks by CO_2 derived from soil. $\delta^{13}C_{DIC}$ values

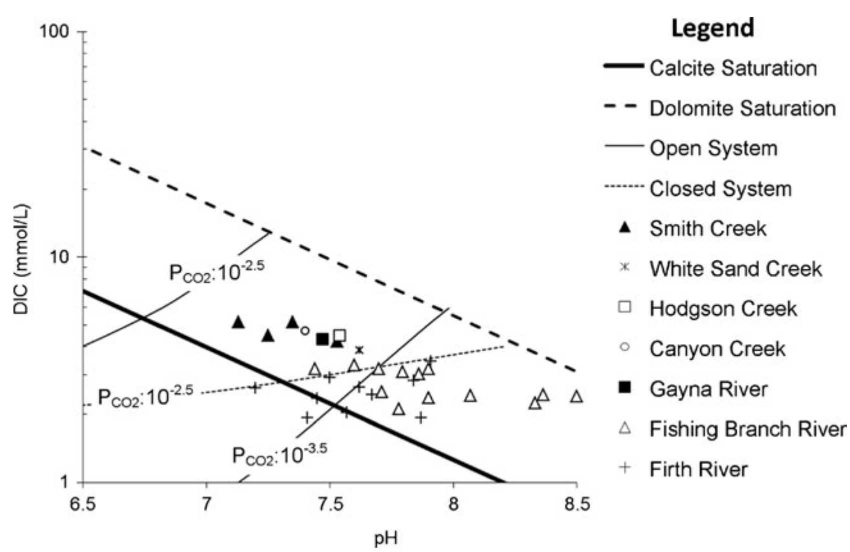

Fig. 4 DIC (mmol/L) vs. pH of water samples plotted with curves for calcite and dolomite saturation as well as open and closed system P_{CO2} evolution curves

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greater than –11 ‰ may be attributed to methanogenesis, as was proposed by Clark and Lauriol (1997) for the Firth River groundwater or by incongruent dissolution. It is expected that methanogenesis would cause the $\delta^{13}C_{DOC}$ to be more enriched than the $\delta^{13}C_{DOC}$ of vegetation derived carbon. The average $\delta^{13}C_{DOC}$ from White Sand Creek, Hodgson Creek, Smith Creek, Canyon Creek, Gibson Creek, Gayna River and the Fishing Branch River is −26.4 ‰ which does not indicate that it has been affected by methanogenesis. Incongruent dissolution of dolomite is supported by the fact that most groundwaters are supersaturated in calcite, but undersaturated in dolomite.

Groundwater recharge and residence time: noble gases

Noble gas concentrations have been measured from groundwater samples collected from springs along the Fishing Branch River, Smith Creek, Gayna River and White Sand Creek (Table 2). These measurements have been used to determine groundwater recharge temperature and groundwater age.

Groundwater recharge temperature

Recharge temperatures of the groundwater springs have been estimated using the inverse modeling approach introduced by Aeschbach-Hertig et al. (1999). The approach constrains temperature and excess air to match modeled dissolved noble gas contents with measured concentrations. Different models exist to describe dissolved noble gases in groundwater as a result of equilibrium dissolution and excess air incorporation (Aeschbach-Hertig and Solomon 2012). The MATLAB program Noble 90 (Peeters et al. 2003) was used to model the noble gas concentrations and to vary the model parameters including temperature, excess air, and fractionation to find the best fit for the observed concentrations. The program minimises χ^2, the error-weighted sum of the model data deviations of the individual noble gases, which at the same time provides a measure of the goodness of the achieved fit (Aeschbach-Hertig et al. 1999). Due to changes in pressure, recharge elevation is a necessary input to constrain the noble gas model and is estimated for each watershed (Table 3). The calculations based on the closed-system equilibration model for dissolved noble gases (Aeschbach-Hertig et al. 2000) return groundwater recharge temperatures between 0±3 and 5±1 °C (Table 3). Groundwater samples from Gayna River and White Sand Creek return recharge temperatures between 0±3 and 1±1 °C, and along the Fishing Branch River between 0±3 and 5±1 °C, which is realistic for these systems. The very high helium concentrations of the Smith Creek sample resulted in problems with measuring the neon concentration, meaning the recharge could not be modelled accurately.

^3H-^3He: groundwater residence time

Helium concentrations and isotope ratios are dependent on the groundwater flow path and groundwater age (Kipfer et al. 2002). Samples from Smith Creek thermal springs had high helium concentrations reflecting a deep groundwater flow path and generally longer residence time. Groundwater from cool springs along Fishing Branch River, White Sand Creek, and Gayna River had lower helium concentrations reflecting a shallower groundwater flow path. Smith Creek groundwater had much higher geogenic helium concentrations due to the deeper flow path and longer residence time of this thermal groundwater. Figure 5 shows $^3He/^4He$ vs. Ne/He of the noble gas samples, where Ne is strictly of atmospheric origin while ^3He and ^4He

Table 2 Noble gas results. *Fishing Branch River* results from Utting et al. (2012), *Smith Creek*, *White Sand Creek* and *Gayna River* results from Utting (2012)

	Lab	S	^4He (cc/g) $\times10^{-7}$	$^3He/^4He$ $\times10^{-6}$	R/R_a	Ne/He	Ne (cc/g) $\times10^{-7}$	Ar (cc/g) $\times10^{-4}$	Kr (cc/g) $\times10^{-7}$	Xe (cc/g) $\times10^{-8}$
Fishing Branch River										
2006 CS 1	H	W	1.53±0.03	1.06±0.01	0.77	1.52	2.32±0.02	4.73±0.02	1.15±0.01	1.66±0.03
2006 Livingstone	O	D		1.14±na	0.83					
2006 S-4	O	D		1.49±na	1.08					
2007 Upper	H	D	1.12±0.02	1.49±0.03	1.08	2.49	2.79±0.02	4.88±0.02	1.12±0.01	1.61±0.04
2007 Upper 2	H	D	1.21±0.02	1.52±0.03	1.1	2.37	2.87±0.02	5.03±0.02	1.15±0.01	1.71±0.04
2007 CS 1	H	W	0.94±0.01	1.02±0.02	0.74	1.52	1.43±0.01	3.85±0.02	1.02±0.01	1.56±0.02
2007 Livingstone	H	D	1.61±0.03	1.16±0.02	0.84	2.01	3.24±0.02	5.43±0.02	1.26±0.01	1.81±0.03
2007 Redd Spring	H	D	1.01±0.02	1.05±0.01	0.76	2.53	2.56±0.02	5.15±0.02	1.21±0.01	1.76±0.03
2007 BC	H	D	8.70±0.02	1.05±0.01	0.76	0.26	2.28±0.02	5.07±0.02	1.23±0.01	1.82±0.04
2007 Gossage	O	D	1.81±0.01	1.37±0.02	0.75	0.99	1.79±0.01			
Smith Creek										
2008-SCW-3.3-HS-P	O	W	15±1	0.0301±0.0004	0.0215	0.01	0.16±0.01	6.2±0.3	1.12±0.06	
White Sand Creek										
2008-WSW-3-S	O	W	1.3±0.1	1.1±0.1	0.783	1.2	1.5±0.1	5.7±0.3		
Gayna River										
2008-GRW-1-S	O	W	1.4±0.1	1.0±0.1	0.736	1.8	2.5±0.2	5.6±0.3	1.04±0.05	
2008-GRW-1-S-Duplicate	O	W	0.88±0.08	1.1±0.1	0.789	2.0	1.8±0.2	6.8±0.3	1.18±0.06	

H stands for samples analysed in Heidelberg and *O* for samples analysed in Ottawa; *S* is the sampler type where *D* is for diffusion and *W* is for water; *R/R_a* $^3He/^4He_{sample}/^3He/^4He_{air}$; *Upper Spring* and *Upper Spring 2* are two different spring vents

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Table 3 Results of noble gas modeling using Noble90 using closed system equilibrium model and age calculations

	Groundwater temperature (°C)	Estimated recharge elevation (ma.s.l.)	χ^2	Noble gas recharge temperature (°C)	ΔNe %	A (ccSTP/g)	F	^{3}H (TU)	^{3}H-^{3}He age (years)
Fishing Branch River									
2006 CS 1	5.6	700	2.82	5±1	19	0.8±1	0.84±0.01	14.3	10±1
2006 Livingstone	4.2							16.3	
2006 S-4	2.3							10.7	
2007 Upper	4.1	700	0.16	3.4±0.7	41	0.023±0.006	0.57±0.033	13.8	16±1
2007 Upper 2	4.5	700	0.97	2.5±0.5	43	0.023±0.005	0.56±0.03	14.1	18±1
2007 CS 1	6.5	700	6.59	1.7±0.4	−29	0.005±0.002	2.4±0.4	13.8	7±1
2007 Livingstone	4.3	700	2.4	0.4±0.4	58	0.02±0.003	0.43±0.03	14.2	11±2
2007 Redd Spring	7.2	700	0.02	2±1	27	0.11±0.08	0.76±0.01	14.8	0.7±1
2007 BC	5.6	700	9.63	0±3	11	0.3±0.9	0.89±0.02	12.3	0±2
2007 Gossage		700						15.5	
Smith Creek									
2008-SCW-3.3-HS-P	10.3	1,000						<0.8	>60
White Sand Creek									
2008-WSW-3-S	3.8	1,500	34.3	0±3	0	0.0±0.9		8.6	30±3
Gayna River									
2008-GRW-1-S	3.1	700	12.4	1±1	31	3.5±1.3		9.2	24±2
2008-GRW-1-S-Duplicate	3.1	700	47.2	0±2	0	0.0±0.9		9.2	20±2

Definitions: χ^2 (Chi squared) represents the error-weighted sum of the model, Δ*Ne* is the amount of excess neon, *A* is entrapped air and *F* is fractionation

may have a geogenic component from U and Th decay and ^{3}He from ^{3}H decay. The high proportion of crustal helium from Smith Creek groundwater is evident by the very low Ne/He. Smith Creek water had tritium concentrations below the detection limit (<0.8 TU) and a ^{3}He/^{4}He of $3.01\pm0.14\times10^{-8}$ which is close to the average crustal value (~2.0×10^{-8}; Kipfer et al. 2002). The ^{3}He/^{4}He of non-thermal groundwater samples ranged from 1.0×10^{-6} to 1.49×10^{-6} and tritium concentrations ranged from 9.2 to 16.3 TU. The concentrations of helium isotopes, neon and tritium were used to determine the groundwater residence time of samples with measureable tritium.

The age of groundwaters which have recharged since the atmospheric testing of nuclear weapons can be determined using ^{3}H-^{3}He dating. The amount of tritogenic ^{3}He from ^{3}H decay is determined by accounting for other sources of ^{3}He such as excess air and crustal helium. This is calculated in Eq. 1 (Kipfer et al. 2002):

$$^3\mathrm{He_{Tri}} = {}^4\mathrm{He_m}(R_\mathrm{m} - R_\mathrm{ter}) - {}^4\mathrm{He_{eq}}\left(R_\mathrm{eq} - R_\mathrm{ter}\right) - L_\mathrm{ex}\left(\mathrm{Ne_m} - \mathrm{Ne_{eq}}\right)\left(R_\mathrm{ex} - R_\mathrm{ter}\right) \quad (1)$$

where ^4He_m is the measured concentration of ^{4}He, R_m is the measured ^{3}He/^{4}He, R_{eq} is the ^{3}He/^{4}He for atmospheric equilibrium (1.36×10^{-6}), R_{ter} is the ^{3}He/^{4}He from crustal sources, $^4He_{eq}$ is the helium concentration in air equilibrated water, L_{ex} is the He/Ne of the excess air component (2.88×10^{-1}), Ne_m is the measured neon, Ne_{eq} is the equilibrium concentration of neon at recharge and R_{ex} is the ^{3}He/^{4}He for excess air (1.384×10^{-6}), (Kipfer et al. 2002). For the groundwaters from White Sand Creek and Gayna River, the ^{3}He/^{4}He value from Smith Creek (3.01×10^{-8}) was used as the crustal value for R_{ter} for groundwater in this region as this groundwater system contained no measurable tritium and is considered close to a purely crustal signal. R_{ter} for groundwater in the Fishing Branch River watershed was determined by regression between air equilibrated water through the data point which obtains the lowest value for R_{ter} on a plot of ^{3}He/^{4}He vs. Ne/He. The value determined for the Fishing Branch River was 6.25×10^{-7}. The value of $^3He_{Tri}$ obtained from Eq. 1 is used in Eq. 2 (Tolstikhin and Kamenskiy 1969) to determine groundwater residence time:

$$t = \frac{1}{\lambda}\ln\left(1 + \frac{^3\mathrm{He_{Tri}}}{^3\mathrm{H}}\right) = \frac{12.3}{\ln 2}\cdot\ln\left(1 + \frac{^3\mathrm{Het_{Tri}}}{^3\mathrm{H}}\right) \quad (2)$$

where 12.3 years is the half-life of tritium, $^3He_{Tri}$ is from tritium decay, and ^{3}H is tritium in water. Results of Eq. (2) are listed in Table 3.

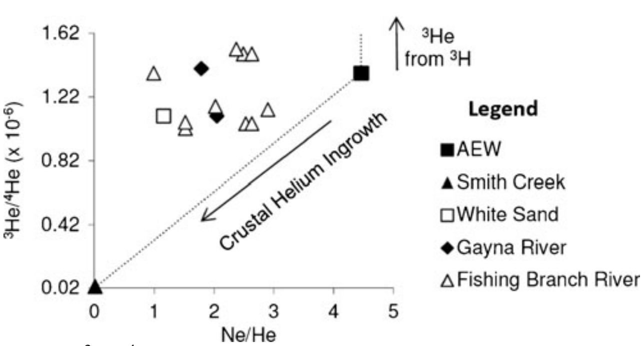

Fig. 5 ^{3}He/^{4}He vs. Ne/He of noble gas samples. *AEW* is air-equilibrated water while *^{3}He from ^{3}H* is the signal of in-growth if the addition of helium is only from tritium. Gas ratios follow the crustal helium in-growth line if only crustal sources of helium are added

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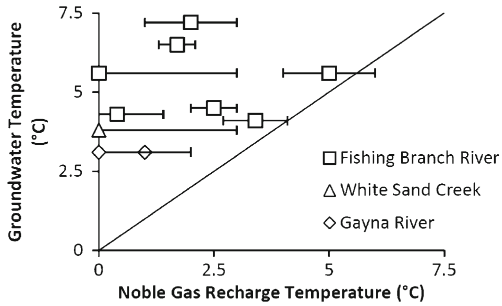

Fig. 6 Groundwater temperature (°C) vs. noble gas recharge temperature (°C) with 1:1 trend line

Groundwater circulation rates are highly variable for the different flow systems. Smith Creek groundwater has no measurable tritium (< 0.8 TU) and a crustal helium signature. This water recharged prior to the 1950s and is classified as submodern. The non-thermal groundwaters contain less crustal helium and have been dated successfully with the ^3H-^3He method. Groundwater along the Fishing Branch River ranges in age from 0±2 to 18±1 years. Groundwater sampled along White Sand Creek was determined to have a groundwater age of 30±3 years. Groundwater sampled along Gayna River was dated at 24±2 years (duplicate sample result 21±2 years).

Discussion

Within the watersheds studied, the measurements of δD, $\delta^{18}O$, $\delta^{13}C_{DIC}$, Ne, Ar, Kr and Xe were used to provide information on groundwater recharge. As δD and $\delta^{18}O$ are dependent on temperature, they provide insights to recharge elevation and potentially the seasonality of recharge. Seven of the groundwater systems studied indicate some degree of depletion of $\delta^{18}O$ and δD. Although depleted isotopic values could be the result of recharge dominated by snowmelt, these values have been interpreted to be the result of recharge at higher elevation with a mix of precipitation through the year. This interpretation is based on the DIC and $\delta^{13}C_{DIC}$ results which support recharge through organic rich soil. Recharging water is a blend of snowmelt and summer rains.

Measurements of $\delta^{13}C_{DIC}$ were used to determine if recharge occurs in organic soils and if it is under open or closed system conditions. The average P_{CO2} of groundwaters is $10^{-2.3}$ atm with the CO_2 derived from organic soils. The $\delta^{13}C_{DIC}$ of groundwater ranges from −3.3 to −12.8 ‰. This range of $\delta^{13}C_{DIC}$ is interpreted to be derived from the dissolution of soil CO_2 and the subsequent weathering of marine carbonates under closed system conditions likely with incongruent dissolution of dolomite affecting some samples. The incongruent dissolution of dolomite along the flow path is supported by the supersaturation with calcite, but undersaturation in dolomite. These processes occur in a similar way in non-permafrost watersheds (Clark and Fritz 1997).

Figure 6 shows the groundwater temperature vs. noble gas recharge temperature. In general, the noble gas recharge temperature is significantly lower than the groundwater temperature at discharge ($P < 0.01$). Groundwaters along White Sand Creek and Gayna Creek, like groundwater from the Fishing Branch River (Utting et al. 2012), have noble gas recharge temperatures that are some 2–3 °C lower than the measured discharge temperatures, signifying no significant advective flux of heat into the subsurface. By contrast, the higher discharge temperatures suggest groundwaters gain heat from elevated ground temperatures in the low-elevation discharge area. Thermal groundwater systems like Smith Creek must gain significant heat in the deeper subsurface to reach >10 °C.

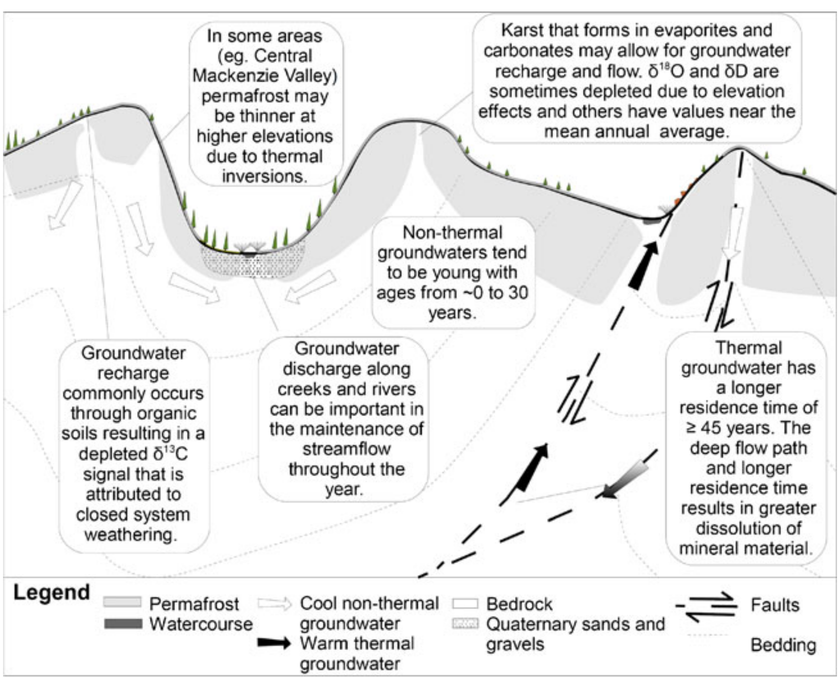

Fig. 7 Schematic diagram of the groundwater flow system

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Based on the noble gas results and the δD, $\delta^{18}O$ and $\delta^{13}C_{DIC}$ of groundwater, it is proposed that groundwater recharge occurs through soil, often in the upper elevations of these watersheds. Six of seven watersheds in the Mackenzie Valley possess groundwater with some isotopic depletion, which suggests recharge at higher elevations. Recharge at these elevations may, to some degree, reflect permafrost distribution as affected by thermal inversions. In the Central Mackenzie Valley, thermal inversions are common in winter (Eley 1974). Under normal conditions it is coldest at higher elevations; however, during inversions this is reversed, potentially resulting in thinner permafrost just below the tree line, to become thicker again at higher elevations, as noted in the Franklin Mountains on the east side of the Mackenzie River (~550 m a.s.l.; Taylor et al. 1998). If zones of thinner permafrost exist just below the tree line, they may act as recharge zones. Recharge water appears to represent precipitation from the entire year. However, recharge may be from variable elevations. Michel (1977, 1986) proposed that recharge may occur from draining of ephemeral lakes underlain by karst in the Franklin Mountains; however, this is not consistent with the $\delta^{13}C_{DIC}$ values presented here. There is abundant karst in the Franklin Mountains and there is likely recharge in karst; however, $^{13}C_{DIC}$ values indicate recharge through organic soils. Recharge must happen during the summer when soils are producing CO_2, and when groundwater temperatures are >0 °C; this water may subsequently flow into karst. The same observations hold true for the Firth River and Fishing Branch River watersheds; however, there was no observable elevation effect.

All groundwater samples had tritium concentrations indicating modern recharge except for Smith Creek groundwater (<0.8 TU). Previous research also found Smith Creek groundwater to have low tritium (7±8, 16 ±8 TU, Michel 1977). The very low tritium concentrations of Smith Creek groundwater indicate groundwater recharge greater than 45 years ago. A small amount of tritium may have been present in early samples due to minor contamination with meteoric water, which in the 1970s had a much higher concentration. Non-thermal groundwaters had groundwater ages ranging from 0±2 to 31±3 years. The ages of non-thermal groundwaters (generally greater than 1 year) suggest that groundwater discharge is probably relatively constant from year to year.

Summary and conclusion

Figure 7 presents a conceptual diagram of the flow system derived from the above results. Despite permafrost conditions, perennial groundwater flow occurs primarily through karst carbonate and evaporite bedrock in Canada's western Arctic. The discharge of this perennial groundwater is manifested in the form of icings and open water year round. Groundwater has $\delta^{18}O$ and δD levels that are either close to the value of mean annual precipitation or are in some cases more depleted. These results suggest that recharge is a mix of annual precipitation and where values are depleted recharge likely occurs at higher elevations. In some areas, permafrost at higher elevations may be thinner than expected due to temperature inversions in winter; this thinner permafrost may allow for enhance groundwater recharge. P_{CO2} ($10^{-2.3}$ atm) and $\delta^{13}C$ (−3.3 to −12.8 ‰) indicate a biogenic supply of CO_2 which is consumed during closed system weathering of marine carbonate material. As such recharge likely occurs through organic-rich soils which overlay fractured bedrock, the weathering of carbonate bedrock results in groundwaters which are often supersaturated in calcite. Noble gas results indicate groundwater recharge temperatures are close to 0 °C, and give no evidence for significant advection of heat into the subsurface during recharge. Groundwater ages are related to groundwater flow path. Non-thermal groundwaters dated by ^{3}He-ingrowth from tritium were found to have circulation times largely on the order of two to three decades, and so an indication that these karst groundwater systems have substantial flow paths and significant storage. Thermal groundwater along Smith Creek is classified as sub-modern based on its lack of measureable tritium.

Isotopic methods suggest that recharge processes in permafrost terrain are not unlike those in non-permafrost terrain. Permafrost reduces the amount of recharge, but in the watersheds studied does not preclude it. The more extensive the permafrost, the more groundwater flow is concentrated in macro-porosity. In other watersheds, even with non-carbonate bedrock, groundwater may flow perennially if there are the appropriate conduits to concentrate flow and heat to prevent freezing.

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