Elsevier Editorial System(tm) for Journal of Environmental Radioactivity

Manuscript Draft

Manuscript Number: JENVRAD-D-16-00246R1

Title: Exploring 222Rn as a tool for tracing groundwater inflows from eskers and moraines into slope peatlands of the Amos region of Quebec, Canada

Article Type: Research Paper

Keywords: peatland; esker; moraine; 222Rn; groundwater discharge; Québec

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Exploring ^{222}Rn as a tool for tracing groundwater inflows from eskers and moraines into slope peatlands of the Amos region of Quebec, Canada

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Highlights

- Radon measured in eskers and connected peatlands in Quebec
- Radon is tested as novel tracer of groundwater inflow in peatlands
- Shallower peat water is depleted in radon by degassing to atmosphere
- Radon and TDS relation suggests inflow from esker and short residence time

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Abstract

Peatlands can play an important role in the hydrological dynamics of a watershed. However, interactions between groundwater and peat water remain poorly understood. Here, we present results of an exploratory study destined to test radon (^{222}Rn) as a potential tracer of groundwater inflows from fluvioglacial landform aquifers to slope peatlands in the Amos region of Quebec, Canada. ^{222}Rn occurs in groundwater but is expected to be absent from peat water because of its rapid degassing to the atmosphere. Any ^{222}Rn activity detected in peat water should therefore derive from groundwater inflow. ^{222}Rn activity was measured in groundwater from municipal, domestic wells and newly drilled and instrumented piezometers from the Saint-Mathieu-Berry and Barraute eskers (n=9), from the Harricana Moraine (n=4), and from the fractured bedrock (n=3). Forty measurements of ^{222}Rn activity were made from piezometers installed in five slope peatlands, along six transects oriented perpendicular to the fluvioglacial deposits. The relationship between ^{222}Rn and total dissolved solids (TDS) measured in water from the mineral deposits underlying the peat layer suggests that ^{222}Rn is introduced by lateral inflow from eskers and moraine together with salinity. This input is then diluted by peat water, depleted in both TDS and ^{222}Rn. The fact that a relationship between TDS and ^{222}Rn is visible calls for a continuous inflow of groundwater from lateral eskers/moraines, being ^{222}Rn rapidly removed from the system by radioactive decay. Although more research is required to improve the sampling and tracing techniques, this work shows the potential of ^{222}Rn tracer to identify groundwater inflow areas from granular aquifers found in eskers and moraines to slope peatlands.

1. INTRODUCTION

Eskers (long, winding ridges of stratified sand and gravel) and moraines (accumulations of unconsolidated glacial debris) are common deglacial morphological features of the northern hemisphere (Clark and Walder, 1994). They can be sites of unconfined to confined granular aquifers, containing extensive reserves of excellent quality groundwater. Their hydrodynamics are still poorly understood, mainly because of the spatial complexity and heterogeneous nature of the fluvioglacial deposits (Boulton et al. 1995).

Peatlands are wetlands with a thick water-logged organic layer composed of living, dead and decaying plant material. Ombrotrophic peatlands (bogs) are peatlands that are almost exclusively rain-fed and that have a higher water table isolated from the regional groundwater flow. In contrast, fens are usually fed by mineral-rich surface water or groundwater. Slope bogs are bogs that have developed on a sloping mineral bedrock or on sloping sediments (defined in NWWG, National Wetlands Working Group 1997). Due to their geomorphic position, slope bogs are often surrounded by marginal fens which can be strongly connected to surface aquifers and can receive water volumes that are not negligible for the peatland water budget or for the entire ecosystem (Glaser et al. 1997). The peat system formed by a slope bog and its marginal fen is hereafter named slope peatland. It is important to quantify groundwater input to slope peatlands in order to understand the hydrodynamics of these environments, but this flux is difficult to measure. Existing studies are mainly based on numerical modeling, water level measurements and water chemistry data (e.g., Ferone and DeVito 2004; Price et al. 2005; Ferlatte et al. 2015; Larocque et al. 2016).

Radon (^{222}Rn) is a noble gas produced by the radioactive decay of ^{226}Ra adsorbed on soil grains or contained in minerals composing the aquifers (Cecil and Green, 2000). The produced ^{222}Rn is then released into the interstitial fluids by α-recoil, diffusion or weathering (Nazaroff, 1992). The ^{222}Rn in turn decays to ^{218}Po, with a half-life of 3.8235 days. The ^{222}Rn content

reaches secular equilibrium (i.e., its quantity remains constant because its production rate equals its decay rate) after a groundwater residence time of approximately 25 days (Andrews et al. 1985). U-rich rocks from denuded Precambrian crystalline basements and composing eskers and moraines are known to produce ^{222}Rn (Ek and Ek 1996; Breitner et al. 2008; Breitner et al. 2010), which migrates into groundwater (Morland et al. 1998; Frengstad et al. 2000; Grolander 2009).

^{222}Rn has successfully been used in hydrology to trace fluid exchange between aquifers and surface water reservoirs such as streams (Hoehn et al. 1992; Ellins et al. 1990; Genereux and Hemond, 1990; Cook et al. 2003; Smith et al. 2012; Lefebvre et al. 2015), lakes (Kluge et al. 2007; Dugan et al. 2011), estuaries (Xu et al. 2014), coastal lagoons (Baudron et al. 2015), and wetlands (Cook et al. 2008; Rodellas et al. 2012). Groundwater is typically enriched in ^{222}Rn compared to surface water. The latter is depleted of ^{222}Rn because of its rapid degassing to the atmosphere. When groundwater discharges into a coastal ocean, lake or stream, ^{222}Rn activity in the surface water reservoir increases to detectable levels. Coupled with measured changes in salinity, electric conductivity or other tracers, ^{222}Rn measurements can be used to quantify these water exchanges (e.g., Stieglitz et al., 2010).

Because peatlands are in direct contact with the atmosphere and there is no internal source of radon within the organic deposits, it is expected that they do not contain ^{222}Rn unless it is introduced via groundwater inflow. The esker-peatland system can therefore be considered comparable to the groundwater-river systems, and it is hypothesized that water exchanges can be traced using ^{222}Rn. The objective of this work was to test ^{222}Rn as a tracer of water flow into eskers and moraines and of inflow from eskers and moraines to slope peatlands. The research was performed in the Abitibi region of southern Quebec (Canada) which has one of the highest densities of eskers in the world (Veillette et al. 2003).

2. STUDY AREA

The study area (860 km^2; Fig. 1a) is located in the Abitibi region of Quebec, Canada. Precambrian rocks forming the bedrock aquifer in the study area are mainly Archean mafic-intermediate volcanic rocks and metasediments of the Superior Province. These lithologies are intruded by Archean granites and tonalites and crosscut by gneissic rocks (Weber and Latulippe 1964). The bedrock aquifer is characterized by low hydraulic conductivity, for which the water bearing potential increases locally with fractures (Cloutier et al. 2007).

The Laurentide Ice Sheet retreat and deglaciation started in the Abitibi region at ~ 9 ka (Dyke 2004) exposing the Precambrian basement and depositing fluvioglacial sediments to form a regional landform, the Harricana interlobate moraine, along with a very dense network of satellite eskers (Veillette et al. 2003). Eskers and moraines are composed of chaotic accumulations of pebble, gravel and sand derived from the glacial erosion of the local Archean bedrock (Veillette et al. 2004). In the study area, three major fluvioglacial landforms are found from west to east (Fig. 1a): Saint-Mathieu-Berry (SMB) esker, the Harricana Moraine and the Barraute esker. The SMB esker is 120 km long, 25 to 45 m thick, and 1 to 5 km wide; the Harricana Moraine has a total length of 278 km and a maximum width of 4.4 km; the Barraute esker is about 20 km long, and is buried over most of its length (Fig. 1a). All of them constitute major granular aquifers (Veillette et al. 2004).

The formation of the proglacial Barlow-Ojibway Lake, and its subsequent drainage towards the Tyrrell Sea 8,200 years ago (Roy et al. 2011), left behind a thick accumulation of fine-grained glacio-lacustrine sediments, named the Ojibway clay. These sediments belt the flanks of the SMB esker and the Harricana Moraine, and created a confining cover on the buried Barraute esker. Direct recharge occurs in unconfined aquifers of the SMB esker and Harricana Moraine. Veillette et al. (2007) and Boucher et al. (2015) suggested that the southern end of the Harricana Moraine (Mont Video; Fig. 1a) could be a recharge area for the buried Barraute esker.

121 Groundwater flows rapidly within the highly porous (average porosity 25-30%; Cloutier
122 et al. 2013) and permeable gravel aquifers formed by eskers and moraines. Groundwater in
123 eskers is mainly of Ca-HCO_3 type, with average salinity of 87 mg/L (Castelli et al. 2011).
124 Groundwater from the fractured bedrock aquifer shows higher salinity up to 760 mg/L (Table
125 1). $^3H/^3He$ water ages in the SMB esker and in the Harricana Moraine range from 6 to 32 yrs
126 (Boucher et al. 2015). For the fractured bedrock, groundwater residence times span from
127 1,473±300 yrs to 137±28 kyrs, suggesting the occurrence of several generations of fossil melt
128 water trapped under the clay plain following the last two glaciations (Boucher et al. 2015).
129 Slope peatlands started accumulating organic matter on esker and moraine flanks after
130 the drainage of the Ojibway Lake, as indicated by ^{14}C dating of basal peat ranging from 8,610
131 to 5,620 yrs (Ferlatte, 2014). The present work focuses on five slope peatlands (Fig. 1a):
132 Sources Nord (SN), Sources Sud (SS), and La Coupe (LC) which border the western flank of
133 the Harricana Moraine, La Belle (LB1, LB2) on the eastern flank of the Harricana Moraine,
134 and Saint-Mathieu-Berry (SMB) located on the eastern flank of the SMB esker. Sandy deposits
135 or clay are found next to the organic deposits and sometimes immediately underneath, forming
136 lateral extensions of local or regional features upon which the organic material has expanded
137 (hereafter called underlying mineral deposits). The peatlands show a marginal fen at their up-
138 gradient limit and fingering organic deposits that drain naturally in small streams at the outer
139 peatland edge (Ferlatte et al. 2015). A regional aquifer characterization study (Cloutier et al.
140 2013) has shown that sand thickness can vary from 1 to 20 m at the peatland border (near the
141 surface aquifer), allowing lateral groundwater – peatland exchanges (Fig. 1b). Using spatio-
142 temporal variations in water levels, Ferlatte et al. (2015) identified two lateral flow patterns at
143 the margin of the slope peatlands (see figures 4 and 5 in Ferlatte et al. 2015 for detailed
144 piezometric heads in all the transects and for typical lateral connections). In some peatlands
145 (LB2, SMB, SN and SS), groundwater from the surface aquifer flows into the peatland and

peatland water flows in the same direction. In the other peatlands (LB1 and LC), water flows from a piezometric mound at station no.2 both into the surface aquifer and towards the peatland center. Vertical hydraulic gradients measured by Ferlatte et al. (2015) suggest that water generally flows downward, i.e., from the peatland to the underlying mineral deposits. Sporadic occurrences of vertical inflows were observed at LB2, SMB and SS (cf. Table 3 in Ferlatte et al. 2015).

3. METHODS

A total of sixteen groundwater samples (Fig. 1a; Table 1) were collected in the study area during the summer of 2012. Samples were collected from the Landrienne municipal well and from two domestic wells in the fractured bedrock aquifer underlying the clay plain (101CH and 55HB; Fig. 1a). Two wells drilled by the Quebec Ministry of Environment (MDDEP-S and MDDEP-P), one piezometer drilled by the *Université du Québec en Abitibi-Témiscamingue* (UQAT; PACES 1-03) and one domestic well at the Mont Video camping site were sampled from the Harricana Moraine. The Barraute municipal well was sampled from the Barraute esker. The Amos municipal well and seven piezometers (TSAM-P1, P2, TSSM-P1, P3, P5, PACES 1-12, 1-13) drilled and instrumented by UQAT were sampled from the SMB esker. Domestic (55HB and 101CH) and municipal (Landrienne) wells in the clay plain have steel casings to the bedrock, and are open from the fractured rock aquifer to the bottom of the well. All other drilled piezometers have a casing and short screens (1.5 m long, except for well TSSM-P5 (3 m screen) and Barraute (4.6 m screen)). The Amos municipal well is a multi-drain well with casing, consisting of eight horizontal drains extending from the base of the vertical borehole with variable lengths of between 7.2 and 45.8 m and a diameter of 0.2 m. Screens cover between 70 and 90% of the horizontal drain length.

In 2010, five peatlands (Fig. 1a) were instrumented with transects of six piezometric stations (Fig. 1b). In each peatland, one or two transects were positioned perpendicular to the peatland margin, in the direction of potential lateral groundwater flow. Transects LB1 and LB2 were located 690 m apart and parallel to each other. For each transect, one piezometer was installed in the surface aquifer (station no.1) while nests of two piezometers were installed in the peatland (stations no.2 to 6). The nests were composed of a surface piezometer (referred to as "(p)" for peat), at a maximum depth of 1.1 m in the peat, and a deep piezometer, 40 cm below the mineral-peat interface (slotted section located in the underlying mineral deposits; referred to as "(m)" for mineral). Each piezometer is made up of a 2.5 cm-ID-PVC pipe, slotted over the lower 30 cm and sealed at the base. In the shallow aquifer (station no. 1; Fig. 1b), the piezometers were installed using an auger and sufficiently deep to ensure that they reach the water table. The shallow piezometer was located within 10 m of the peatland margin; station no.2 is located as close as possible to the peatland margin, where peat thickness reaches 40 cm; station no.3 is located where significant changes in vegetation are observed (transition from a fen vegetation to a bog vegetation); stations no.4, 5, and 6 are approximately 50, 100 and 300 m away from station no.3 respectively (Fig. 1b) (Ferlatte et al., 2015). Major ions and trace elements were measured from water sampled at each of the stations (Ferlatte, 2014; Larocque et al. 2016). Ferlatte et al. (2015) used slug tests in temporary piezometers to measure peat hydraulic conductivities between stations no.2 and 3. Their results showed that hydraulic conductivities decrease with depth and for the SMB peatland vary between $5.5x10^{-6}$ $m.s^{-1}$ at 0.50-0.75 m to $6.9x10^{-7}$ $m.s^{-1}$ at 1.00-1.25 m below the surface.

In the municipal wells (Amos, Landrienne, and Barraute), groundwater was collected directly at a faucet connected directly to the wellhead. Groundwater from the monitoring wells and the peatland piezometers was collected using a Waterra® Inertial Pump System which consists of a foot valve located at the bottom of a high-density polyethylene tube with a variable

195 diameter of 5/8" to 1" and an electric actuator Hydrolift-2® pump. Groundwater from domestic
196 wells (55HB, 101CH, Mont Video) was collected at the water faucet closest to the well, outside
197 the house to avoid home reservoirs where the water undergoes degassing. Water was purged
198 from monitoring wells until electrical conductivity, pH, and temperature stabilized. Peatland
199 piezometers were purged to ensure the collection of fresh water. Sampling was done three days
200 following the purge to allow time for water level recovery and thus to have a sufficient volume
201 of water to be recovered in our sampling vessels. A volume of water equivalent to the length of
202 the piezometer was purged prior of sampling to be sure that no stagnant degassed water was
203 collected. Radon was collected using 250 cm^3 glass bottles by inserting a PVC tube to the
204 bottom. We ensured a constant and low flux of water pumped by the Waterra® to avoid water
205 agitation and bubbling, which could provoke radon degassing. Once filled, the bottle was
206 rapidly sealed with a plastic cap and examined visually for the presence of any air bubble
207 (Leaney and Herczeg, 2006). The bottles were labeled with the time of sampling in order to
208 correct the measured ^{222}Rn activities of the time lap between field collection and laboratory
209 analyses.

210 Only 40 peatland piezometers (of a total of 57) contained sufficient water to allow robust
211 ^{222}Rn analysis. In 12 piezometers, there was enough water to take two consecutive samples for
212 ^{222}Rn analyses. The ^{222}Rn activity was measured with a liquid alpha scintillation counter (Hidex,
213 LS 300) in the GEOTOP laboratories, Université du Québec à Montréal. Scintillometer
214 efficiency was calibrated using international standards. Sampling and analysis protocols were
215 optimized and analytical tests were performed to obtain the best accuracy and precision of ^{222}Rn
216 activity in a continuous interval from 0.5 to 35 Bq/L. Sample preparation for ^{222}Rn measurement
217 was performed using the extraction method (EM; Leaney and Herczeg 2006) as follows: 25
218 cm^3 of scintillant were mixed with the entire volume of sampled water (250 cm^3).
219 Approximately 10 cm^3 of air were introduced into the glass bottle to facilitate emulsion between

220 water and the scintillant. The cocktail was mixed for 4-5 min and left to stand until phases
221 separate. A volume 8 cm^3 of scintillant was then extracted and added to the counting glass vials.
222 During calibration, mean background noise values were smaller than 0.12 Bq/L, with an
223 average error of 4% for the analysis. Data reported here are corrected for the background noise.
224 Details of the sample preparation and analyses are given in Lefebvre et al. (2013) and Pinti et
225 al. (2014).

226 Five water samples (LB2.no2(m), LB2.no3(m), SN.no2(m), SMB.no2(p) and
227 SMB.no3(m); Table A1) contained 10 to 30% suspended particles volumetrically. These
228 particles can contribute to a sustained flux of ^{222}Rn in water through the decay of ^{226}Ra adsorbed
229 on the clay or organic particles. To quantify this flux, the ^{226}Ra activity from the solid phase
230 was measured at the GEOTOP, using a high-purity Germanium well detector gamma-ray
231 spectrometer equipped with an ORTEC® DSPEC jr. 2.0 interface. Eight grams of soil matrix
232 from the five samples were closed and left in vials for at least 25 days to ensure that secular
233 equilibrium between ^{226}Ra and ^{222}Rn was reached. The ^{226}Ra activity was obtained by measuring
234 the peak of ^{214}Pb at 352 keV and 295.2 keV, and the peak of ^{214}Bi at 609keV (see Pickler et al.,
235 2012 for details). Results are reported in appendix (Table A1) together with a discussion on the
236 contribution of ^{226}Ra to peat water samples.

237 Uranium (U) concentration in peatland sediments was calculated from the ^{226}Ra activity
238 at 352 keV and assuming secular equilibrium (1 ppm U = 12.34 Bq/kg ^{226}Ra). The U content in
239 rocks composing the aquifer matrix of the eskers and moraines was measured on selected rock
240 samples collected in a quarry on the top of the Harricana Moraine. The U content was
241 determined by neutron activation at the Slowpoke reactor of the Physics Department of the
242 Université de Montréal. Results are reported also in Table A1 in the appendix.

243 In eight piezometers (LB2.no6(p), SS.no6(p), SMB.no6(p), SN.no2(p), SN.no3(p),
244 SN.no4(p), SN.no5(p), SN.no6(p)), the volume of water available was sufficient to be pumped

through refrigerator-type copper tubes with stainless steel clamps at each extremity while avoiding contact with ambient air (e.g., Boucher et al., 2015). A 14 cm^3 volume of water was trapped in the copper tube and then degassed under vacuum at the Noble Gas Laboratory of the GEOTOP to measure noble gas contents. These analyses were aimed at testing whether we can use atmosphere-derived noble gas isotopes ^{36}Ar, ^{84}Kr and ^{132}Xe dissolved in peatland water as an indicator of water degassing and thus be able to relate radon variability in sampled peatlands to local degassing in the peatland system or to the natural variability of radon in water systems, which is very often observed (e.g., Harris et al. 2006). Water was degassed under vacuum and purified to analyze the single isotopes by using a quadrupole mass spectrometer (QMS) Prisma-200C from Pfeiffer®, equipped with a Faraday cup and a channeltron multiplier. The instrument was calibrated daily with a purified air standard. Analytical details on noble gas measurements are reported in Roulleau et al. (2012). Results are reported in appendix Table A2.

3. RESULTS

^{222}Rn activities measured in esker and moraine groundwater were found to range from 2.8 (PACES 1-13; SMB esker) to 12.5 Bq/L (PACES 1-03; Harricana Moraine) (Table 1). Groundwater from the fractured bedrock aquifer underlying the Ojibway clay plain shows higher ^{222}Rn activity, ranging from 14.7 (101CH) to 34.9 Bq/L (Landrienne municipality well) (Table 1). The Amos municipal well was sampled twice and the obtained ^{222}Rn activities were 11.1 and 11.5 Bq/L. The TDS (Table 1) was calculated as the sum of the measured dissolved major ions, Ca^{2+}, Mg^{2+}, Na^+, K^+, Cl^-, SO_4^{2-}, and HCO_3^-. The TDS is higher in the fractured bedrock (ranging from 351 to 763 mg/L) than in the eskers and moraine aquifers (ranging from 45 mg/L at Mont Video to 251 mg/L in the Barraute municipality well).

The ^{222}Rn activities measured in peat water range from 0.1 (LB1.no5(p)) to 1.0 Bq/L (SMB.no6(p)), while groundwater from the mineral deposits underlying the peatland shows

higher and more variable ^{222}Rn activity, ranging from 0.02 (LB1.no3(m)) to 16.6 Bq/L (SS.no4(m)) (Table 2). One piezometer (LC.no1) was installed on the esker, at the edge of the peatland, and its ^{222}Rn activity (8.1 and 8.8 Bq/L; Table 2) relates to groundwater inflow. Two consecutive water samples were taken from twelve piezometers for ^{222}Rn analyses, when the volume of water in the piezometer was sufficient (Table 2). In general, the second sample shows higher ^{222}Rn activity than the first. This could indicate water stratification in the peatland. Water sampled first might be partially stagnant and degassed of its ^{222}Rn content. The second sample might represent deeper water, isolated from the atmosphere and having preserved the total amount of dissolved ^{222}Rn. As mentioned in the "Methods" section, a third consecutive sample of water was taken into copper tubes to measure the atmospheric noble gas composition whenever there was sufficient water (Appendix). Results (Table A2) show that samples LB2.no6(p) and SMB.no6(p) clearly underwent degassing (16-32% and 36-42% of the total, respectively). For all the other samples, degassing was minimal (2-3%) or practically null (Table A2), indicating that deeper water is less affected by degassing and that the purged volume of water should be increased to avoid the risk of collecting degassed samples (e.g., Harris et al. 2006).

TDS concentrations for peat water and for water in the underlying mineral deposits were calculated as the sum of the dissolved major ions for the May and September 2011 samplings of Ferlatte (2014). With a few exceptions, TDS values in May and September were similar and the average TDS value was considered sufficiently representative to be used henceforth. Average TDS values range from 4.0 to 22.2 mg/L in the peat water and from 10.4 to 41.5 mg/L in the underlying mineral deposits. One outlier (sample SS.no6(m)) shows much higher TDS, with an average concentration of 144.5 mg/L (Table 2).

4. DISCUSSION

295 *4.1 ^{222}Rn activity in eskers and peat water: comparison with similar environments*

296 Very few data of ^{222}Rn activity in groundwater from eskers and moraines exist in
297 literature. Indeed, the majority of the studies are for soil gas radon carried out for environmental
298 and health issues (e.g., Akerblom et al., 1994). Results for the current study are compared to
299 the few data existing from shallower wells tapping groundwater from Quaternary deposits
300 (eskers and moraines) in Scandinavian regions. In these regions, fluvioglacial deposits are
301 derived from the denudation of Archean igneous and metamorphic rocks, similarly to those of
302 the Canadian Precambrian Shield of Abitibi. Fennoscandian fluvioglacial Quaternary deposits
303 are geologically the equivalent of those of Abitibi.

304 The average ^{222}Rn activity of 7.7 Bq L^{-1} in groundwater from eskers and moraine of the
305 Abitibi region are in the lower ends of the values measured in shallow wells from esker-covered
306 Fennoscandian regions (up to 70 Bq/L; Banks et al. 1998; Vesterbacka et al. 2005; Grolander
307 2009; Fig. 2a). The lower ^{222}Rn activity measured in the Amos region esker and moraines are
308 likely related to the lower U content in these rocks compared those from the Fennoscandian
309 shield. U contents are relatively low, between 0.2 to 0.9 ppm as measured in Harricana Moraine
310 sediments (Table A2). Fennoscandian granites and metamorphic rocks are known to contain
311 notable amounts of U (several tenths or hundreds ppm), producing high ^{222}Rn activity
312 (Frendgstad et al. 2000). For example, in deep wells drilled directly in the granitic bedrock of
313 the Fennoscandian craton, measured ^{222}Rn activities are extremely high, with values ranging
314 from 700 to 30,000 Bq/L (Frendgstad et al. 2000). In the Amos region, the maximum ^{222}Rn
315 activity from wells tapping water directly from the crystalline basement is 34.9 Bq/L
316 (Landrienne municipality well; Table 1).

317 In the Amos region, ^{222}Rn activity expected in esker/moraine groundwater or from the
318 crystalline basement can be estimated from the U content of the rock (0.2-0.9 ppm), using the
319 Eqn. A1 in the appendix. However, because it is assumed that the secular equilibrium between

320 $^{226}Ra_{sediment}$ and $^{222}Rn_{water}$ has been reached in the aquifer (t = 25 days; Andrews et al. 1985), the
321 term $1\text{-}e^{\lambda t}$ Eqn. A1 can be ignored. Resulting ^{222}Rn activity in groundwater varies from 1.8 to
322 25.6 Bq/L, in the range of the measured values (Table 1).
323 Because there are no other study of ^{222}Rn in peatlands, measured ^{222}Rn activities in peat
324 water from the current study were compared with those from other wetland environments, such
325 as coastal wetlands and mangrove sites (Cook et al. 2003; Cook et al. 2008; Gleeson et al. 2013;
326 Maher et al. 2013; Zarroca et al. 2013; Fig. 2b). ^{222}Rn activities from these environments can
327 be compared to those from the slope peatlands set on eskers because they are expected to have
328 accumulated some ^{222}Rn, mainly from radium originating in the mineral substratum and from
329 groundwater ^{222}Rn. It is clear that this is a first-order approximation because of the different
330 hydrological dynamics that distinguish these wetland environments. The majority of the
331 shallower peat water samples from this study (designated "p" in Table 2), show ^{222}Rn activities
332 lower than 1 Bq/L, similar to the range of activities measured in the other wetland environments
333 (Fig. 2b). This minimum activity of 1 Bq/L probably translates the equilibrium between ^{222}Rn
334 produced in the aquifer rocks and that degassed to the atmosphere. Water samples from the
335 mineral substratum of the peatland show ^{222}Rn activities significantly higher than reported in
336 the wetlands literature (up to 16.6 Bq L^{-1}) and comparable to those from the eskers or the
337 fractured bedrock (Table 1).
338
339 *4.2 ^{222}Rn and TDS in peatland water as an indicator of water mixing and residence time*
340 ^{222}Rn activities recorded in water from the peat (shallower piezometers "p") has an
341 average^{222}Rn activity of 0.36 Bq/L, while water from the mineral substratum (deeper
342 piezometers "m") has an average ^{222}Rn activity of 4.7 Bq/L (Fig. 2b). The observed difference
343 can be accounted for 1) degassing of radon towards the atmosphere in piezometers located
344 closer to the surface, or 2) dilution from rainwater depleted in ^{222}Rn.

There is a clear trend between the TDS (salinity) and the ^{222}Rn activity measured in the mineral substratum water (black dots; Fig. 3) which is not present for the peat water samples (white circles; Fig. 3). This trend has been observed in studies of groundwater discharge in coastal wetlands (Rodellas et al., 2012) and of coastal aquifer interactions with seawater (e.g., Stieglitz et al., 2010). In the latter case, the observed trend would be the opposite of that observed here, i.e. low salinity, high ^{222}Rn groundwater mixes with high salinity, ^{222}Rn-depleted seawater. In order to observe a trend between a conservative tracer such as salinity and a non-conservative tracer such as ^{222}Rn (Fig. 3), which is affected by radioactive decay and degassing, the residence time of ^{222}Rn in the wetland must be short, i.e. there has to be a continuous inflow of ^{222}Rn rich groundwater from the eskers/moraines. Longer residence times will lead to a less clear trend because of the radioactive decay of ^{222}Rn. The trend observed in deeper piezometers in the mineral substratum clearly indicates that mixing between two sources of water and ^{222}Rn dominates radioactive decay and degassing (Stieglitz et al., 2010). The mixing would be between high TDS and ^{222}Rn-rich groundwater from eskers/moraines which is progressively diluted by peat water depleted in both TDS and ^{222}Rn (Fig. 3). This mixing implies that the blurring of the TDS-radon relation in the shallower piezometers can be caused only by degassing of ^{222}Rn close to the surface.

Based on the TDS-^{222}Rn relationship (Fig. 3), the TDS of the peat water (i.e. when ^{222}Rn reach zero at the abscissa intercept) is ~12 mg/L which can be interpreted as a threshold below which there is no more ^{222}Rn-rich groundwater inflow into the peatland. This threshold is compatible with the independent calculation of 16 mg/L derived by Larocque et al. (2016) for the peat water using major ions geochemistry. Figure 4 summarizes the possible ^{222}Rn sources and pathways between the eskers and the slope peatlands.

4.3 Limitations and improvements

370 It is clear from the results of this study that much work remains to be done to improve
371 both the sampling techniques, particularly in the peat layer where the water volumes necessary
372 to reliably detect and measure ^{222}Rn are limiting. *In situ* repeated measurements using a portable
373 radon scintillometer would limit the amount of water used and increase the precision of the
374 measurement. The few atmospheric-derived noble gas data obtained in this study suggest that
375 their systematic analyses together with radon will be extremely helpful in determining
376 degassing processes *in situ* in order to understand the measured variability in radon in repeated
377 measurements. Use of passive diffusion samplers, i.e. devices in which gas diffuses through a
378 semi-permeable membrane connected to a small-volume copper tube (e.g., Aeschbach-Hertig
379 and Solomon, 2013) inserted at the bottom of the peatland piezometers might be ideal for
380 measuring with precision the degassing conditions of the water in the peat layer. Finally,
381 dedicated nests of several piezometers inserted into the peat and the underlying mineral deposits
382 at different depths at each location could be useful to measure ^{222}Rn activity gradients in order
383 to quantify flows and determine their dominant directions.
384
385 **5. CONCLUSIONS**
386 The objective of this work was to test ^{222}Rn as a potential tracer of groundwater inflow
387 from eskers and moraine to slope peatlands in the region of Amos, Quebec. The distribution of
388 ^{222}Rn measured in eskers/moraine and in slope peatlands of the Abitibi region shows that its
389 activity in eskers is enriched by on average one order of magnitude compared to that measured
390 within the peatlands, mainly because ^{222}Rn is degassed to the atmosphere in the shallower peat
391 layer. The measured ^{222}Rn activity in the eskers and moraine is compatible with the values
392 expected at secular equilibrium with its progenitor, ^{226}Ra contained in the aquifer rocks. The
393 observed relationship between ^{222}Rn and TDS in the mineral deposits below the peatlands
394 indicate a mixing between two water sources, one enriched in salt and ^{222}Rn which could be

identified as groundwater in flowing into peatlands. This work also highlights the limitation of the method and *in situ* measurements of ^{222}Rn, possibly continuous monitoring of ^{222}Rn and other parameters such as salinity could be the key to quantify groundwater inflows into peatlands.

ACKNOWLEDGEMENTS

This study was funded by the *Fonds de Recherche Québec - Nature et Technologie* (project no. 136990 of the *Programme de recherche en partenariat sur les eaux souterraines du Québec to DLP, MR and ML*) and NSERC Discovery Grant no. 314496 to DLP. The municipalities of Amos, Landrienne and Barraute, as well as D. Blanchette (UQAT), C. Boucher, P. Méjean and G. Vautour (GEOTOP-UQAM) are thanked for their help and cooperation during sampling.

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578 **FIGURE CAPTIONS**

579

580 **Figure 1**. (**a**) Schematic map of the sampled area, with the position of the Saint-Mathieu-Berry
581 (SMB) esker, the Barraute esker and the Harricana Moraine. Black circles are groundwater
582 wells located on the Harricana Moraine, white circles are groundwater wells located on the
583 SMB and Barraute eskers, and grey squares are groundwater wells tapping the fractured
584 bedrock aquifer. Grey zones represent the areal extent of the monitored peatlands with white
585 squares indicating the location of the instrumented transects. (**b**) Schematic profile showing
586 how peatlands were instrumented with piezometers (modified from Ferlatte et al. 2015).

587

588 **Figure 2**. Measured ^{222}Rn activities a) in wells located in esker, moraine and bedrock aquifers,
589 and b) within the peatlands and in the mineral deposits underlying the peatlands, compared with
590 literature values from similar environments. References are: [1] Gronlander 2009; [2]
591 Vesterbacka et al. 2005; [3] Banks et al. 1998; [4] Zarroca et al. 2013; [5] Cook et al. 2008; [6]
592 Gleeson et al. 2013.

593

594 **Figure 3**. ^{222}Rn activity measured in water from the peat layer (white circles) and in the
595 underlying mineral deposits (black circles), plotted against TDS. The TDS is the average of the
596 two measured values (May and September 2011), and the vertical error bars represent the
597 maximum and minimum values (see Table 2 and Ferlatte, 2014). For the piezometers with an
598 asterisk in Table 2, the symbols indicate the average of repeated ^{222}Rn measurements with
599 uncertainties. ^{222}Rn activities of samples SMBno.6(m) and SSno.4(m) are significantly different
600 between repeat measurements, indicating accidental degassing, and the maximum values have
601 instead been reported here. The correlation has been calculated by excluding outliers (one
602 sample: SS.no6(m)), determined as values that are $\geq 1.5\sigma$ larger than the calculated mean value.

603 Arrows indicate how radioactive decay, degassing, mixing and groundwater inflows affect the
604 ^{222}Rn-TDS relationship.
605
606 **Figure 4**. Schematic illustrating the potential ^{222}Rn sources, the processes affecting ^{222}Rn
607 activity and the ^{222}Rn migration paths in the eskers/moraine-peatland systems of Abitibi.
608

609 **Appendix**

610 *The ^{226}Ra and ^{238}U in peat and esker sediments*

611 Table A1 report the γ-measurements carried out in the sediments found in water samples
612 (LB2.no2(m), LB2.no3(m), SN.no2(m), SMB.no2(p) and SMB.no3(m). The ^{226}Ra activities
613 measured in sediments show high variability, with values ranging from 5.04 to 6.07 Bq/kg for
614 sample LB2.no3(m), and from 27.12 to 34.12 Bg/kg for sample SMB.no2(p). ^{226}Ra activities
615 measured in the sandy sediment of LB2.no2(m) and LB2.no3(m) are similar to each other and
616 are the lowest of the five water samples from which sediments could be analyzed. ^{226}Ra activity
617 apparently increases in sediments where till deposits and clay are present. The highest ^{226}Ra
618 activity is found in clay sediments (i.e., sample SMB.no3(m)), where the adsorption capacity
619 of radium is the highest. However, SMB peat water is very acidic (pH = 3.9-4.4; Ferlatte 2014)
620 and the sorption capacity of radium is likely inhibited in this sample, as observed in other acidic
621 environments (Pickler et al., 2012).

622 The ^{226}Ra activities were used to estimate the activity of ^{226}Ra-supported ^{222}Rn in the
623 water samples and the U content of the peatland mineral substratum. Calculated U
624 concentrations in the sediment range from 0.41 (LB2.no2(m)) to 0.82 ppm (SN.no2(m)), with
625 an outlier of 2.52 ppm for SMB.no2(p)) (Table A1).

626 The ^{222}Rn contribution from the suspended sediment to peat water in samples
627 LB2.no2(m), LB2.no3(m), SN.no2(m), SMB.no2(p) and SMB.no3(m) (Table A1) can be
628 calculated following Eqn. A1, modified from Bonotto and Caprioglio (2002):

629

630 $$\left[{}^{222}Rn\right]_{water}(BqL-1) = 12.469 \cdot \rho \cdot \left(1-\phi/\phi\right) \cdot A_{Rn} \cdot \left[U\right]_{rock} \cdot (1-e^{\lambda t}) \qquad \text{(Eqn. A.1)}$$

631

632 where ρ is the bulk density of the rock (g cm^{-3}), ϕ is the porosity and (1-ϕ/ϕ) is the void ratio
633 (unitless), A_{Rn} is the emanation coefficient of ^{222}Rn, [U] is the U concentration in ppm, λ is the

decay constant of ^{222}Rn (0.00755 hr^{-1}) and *t* is the time interval (in hours) between sampling and analysis. The void ratio has been assumed to range between 0.11 and 0.43, corresponding to the 10 to 30% volume of sediment in 250 cm^3 of water contained in the sample. The time interval between sampling and analysis varied between 73 and 233 hours. The emanation coefficient is the fraction of the total amount of ^{222}Rn produced by ^{226}Ra decay that escapes from the rock to the pore water. It is a difficult parameter to evaluate and depends on lithology, temperature and moisture content (Nazaroff 1992). Nazaroff and Nero (1988) suggest values of between 0.06 and 0.18 (an average value of 0.14) for sands. More recent experimental work on granite sand from eskers shows similar emanation coefficients, with values ranging from 0.17 to 0.24 (Breitner et al. 2008) and 0.12 to 0.30 (Breitner et al. 2010).

The ^{222}Rn contributed from the sediment to peat water can range from 1.9 to 8.3% of the total measured ^{222}Rn activity for a sediment volume of 10% and an A_{Rn} of 0.12, and from 18 to 80% of the total measured ^{222}Rn activity for a sediment volume of 30% and an A_{Rn} of 0.30 (Table A1). This calls for caution in taking peat water samples so as to avoid as much as possible the collection of suspended material. Filtration during sampling with a large mesh could reduce the collection of larger sediment particles, though this process may be unfeasible, given the limited amount of available water.

Atmosphere-derived noble gases

Table A2 report the abundances of atmosphere-derived noble gas isotopes ^{36}Ar, ^{84}Kr and ^{132}Xe in cm^3STP (Standard Pressure and Temperature) per gram of water, measured in peatland water samples. These abundances were also reported in the F-notation. In F-notation, measured abundances are normalized to the atmospheric abundance dissolved in groundwater or ASW (Air Saturated Water) with ^{36}Ar as the reference isotope:

659 $F(i)=(i/^{36}Ar)_{sample}/(i/^{36}Ar)_{ASW}$

660

661 where *i* represents ^{84}Kr and ^{132}Xe. ASW noble gas composition is calculated at the sampling
662 temperature and using solubility data in groundwater from Smith and Kennedy (1983). F-values
663 are fractionation factors that provide a measure of the enrichment or depletion relative to the
664 ASW composition and thus they are very useful fingerprints of occurrence of peatland water
665 degassing that could have affected radon data.

Table A1. Gamma-ray measurements of ^{226}Ra activity in peatland sediments and neutron-activation U concentration in Harricana rocks

Piezometer name	Mineral substratum*	^{226}Ra	±	^{226}Ra	±	^{226}Ra	±	^{226}Ra contrib. to ^{222}Rn	[U]**
		295.2 keV ^{214}Pb		352 keV ^{214}Pb		609 keV ^{214}Bi			
		Bq/kg		Bq/kg		Bq/kg		%	ppm
LB2.no2(m)	Sand	5.57	1.01	5.04	0.64	6.76	1.13	3.65-35.19	0.41
LB2.no3(m)	Sand	5.04	0.96	6.29	0.72	6.39	1.11	1.92-18.52	0.51
SN.no2(m)	Sand/Till	11.46	2.04	10.10	1.29	13.79	2.30	8.29-79.93	0.82
SMB.no2(p)	Sand/Clay	34.12	5.34	31.07	3.39	27.12	4.97	-	2.52
SMB.no3(m)	Clay	9.75	1.86	8.91	1.19	11.79	2.13	3.25-31.33	0.72
Harricana	Clay-silt								0.75
Harricana	Clay-silt								0.75
Harricana	Sand								0.36
Harricana	Sand								0.33
Harricana	Arkose								0.20
Harricana	Conglomerate								0.87
Harricana	Volcanic rock								0.84

*: Sediment substratum for peatland are from Ferlatte et al. (2014).
666 **: U concentration in Harricana moraine from Boucher (2013).

667

668
669

670

671
672 Table A2. Measured atmospheric noble gas amounts and calculated fractionation factor "F" in
673 selected peatland piezometers.
674

Well	Temperature	^{36}Ar**	^{84}Kr	^{132}Xe	F(^{84}Kr) ***	F(^{132}Xe)
	(°C)*	$cm^3STP\ g^{-1}_{H2O}$				
LB2.no6(p)	11.3	$1.14x10^{-6}$	$4.29x10^{-8}$	$2.35x10^{-9}$	0.84	0.68
SMB.no6(p)	12.0	$8.32x10^{-7}$	$3.21x10^{-8}$	$1.97x10^{-9}$	0.64	0.58
SN.no2(p)	10.6	$1.53x10^{-6}$	$5.87x10^{-8}$	$3.76x10^{-9}$	1.13	1.06
SN.no3(p)	8.1	$1.71x10^{-6}$	$6.68x10^{-8}$	$4.21x10^{-9}$	1.20	1.09
SN.no4(p)	8.2	$1.43x10^{-6}$	$5.68x10^{-8}$	$3.83x10^{-9}$	1.02	1.00
SN.no5(p)	8.6	$1.13x10^{-6}$	$5.02x10^{-8}$	$4.13x10^{-9}$	0.92	1.09
SN.no6(p)	8.9	$1.24x10^{-6}$	$5.20x10^{-8}$	$3.62x10^{-9}$	0.96	0.97
SS.no6(p)	9.9	$1.07x10^{-6}$	$5.16x10^{-8}$	$3.88x10^{-9}$	0.98	1.07

675 * average water temperature at the site. Data from Ferlatte (2014).
676 ** average uncertainties on the measured isotopic abundances are ±5% of the measured value.
677 *** calculated uncertainties on the F(i) values are 7.5% of the calculated value.
678

Figure 1

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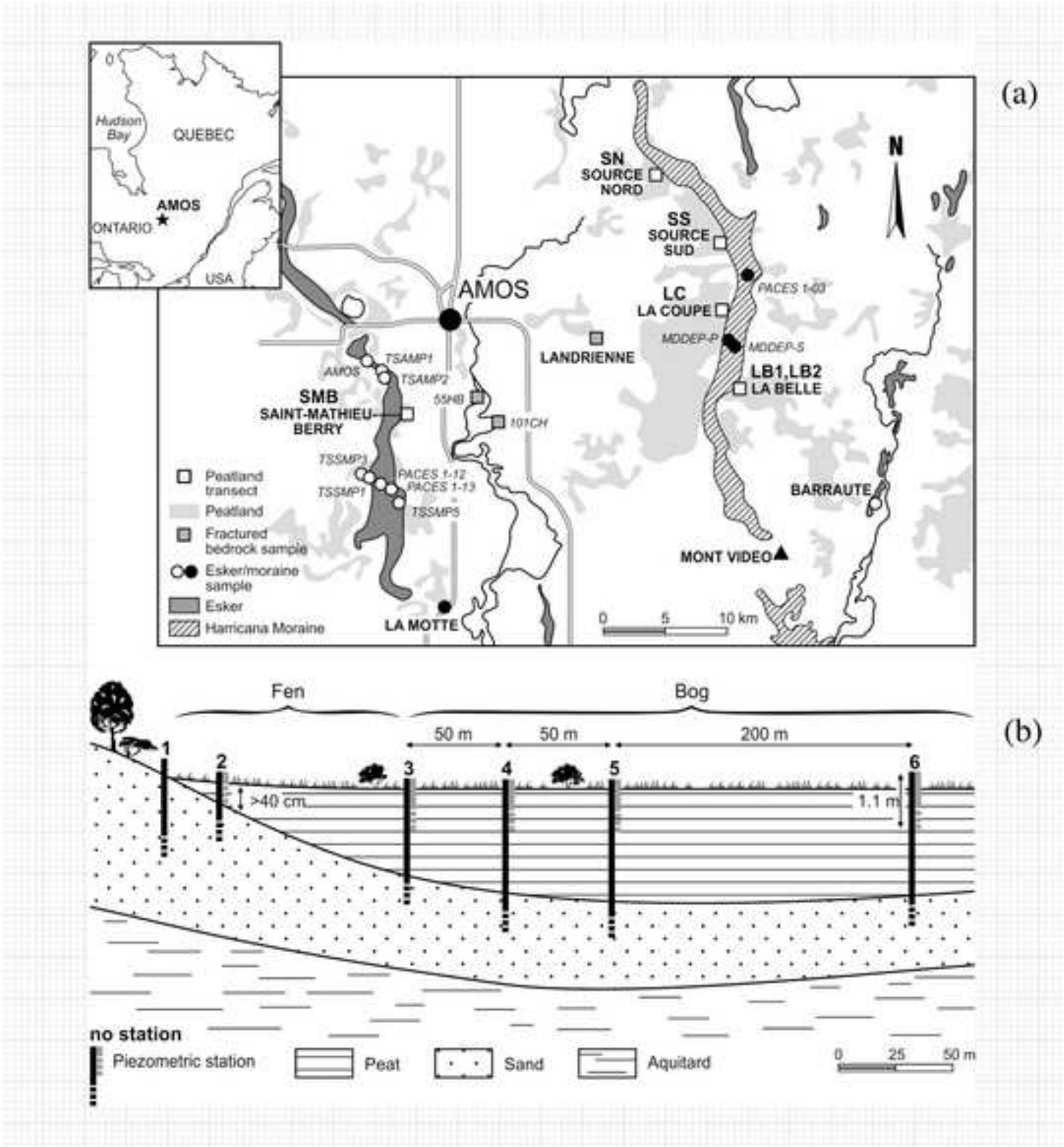

Figure 2

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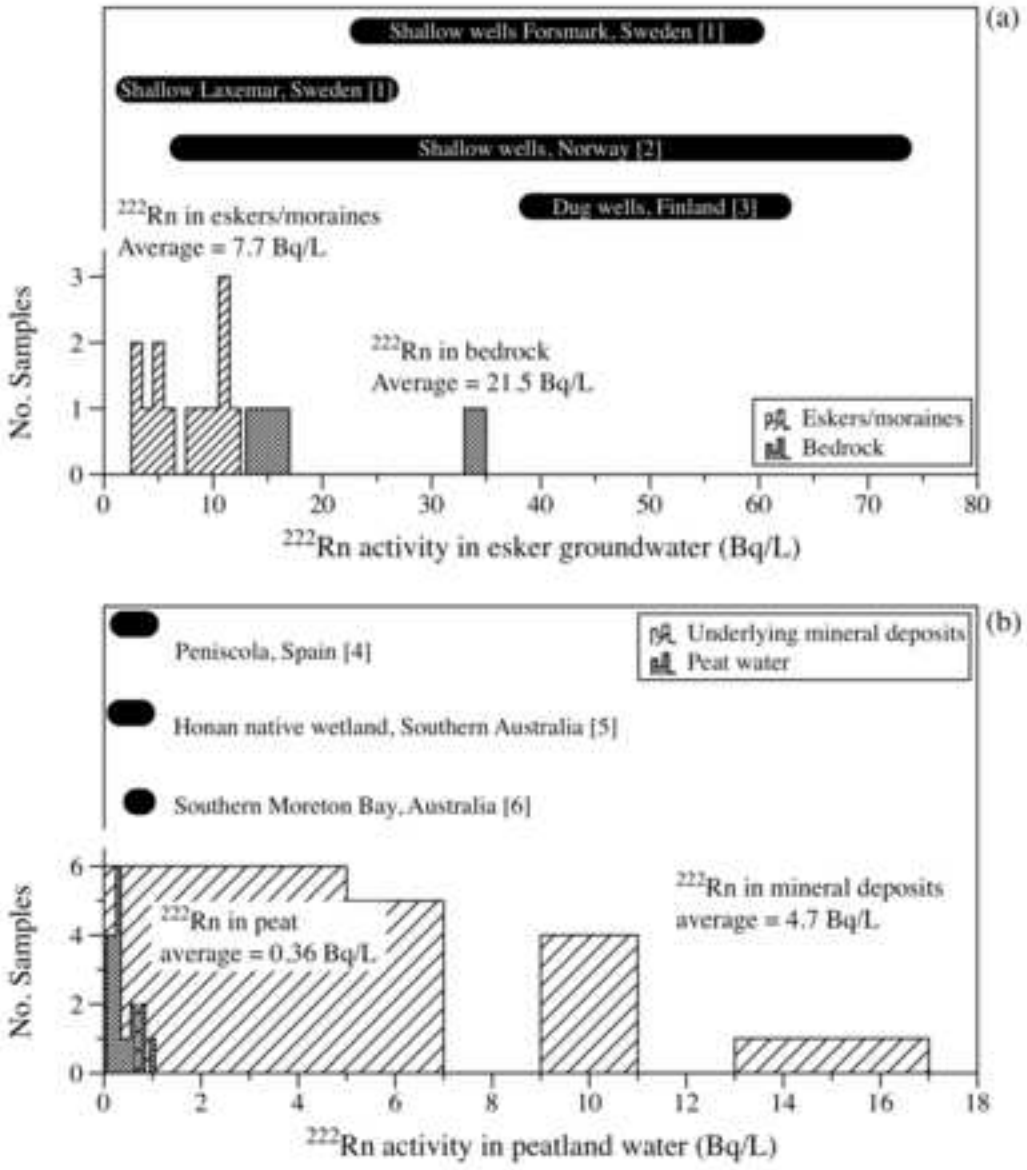

Figure 3

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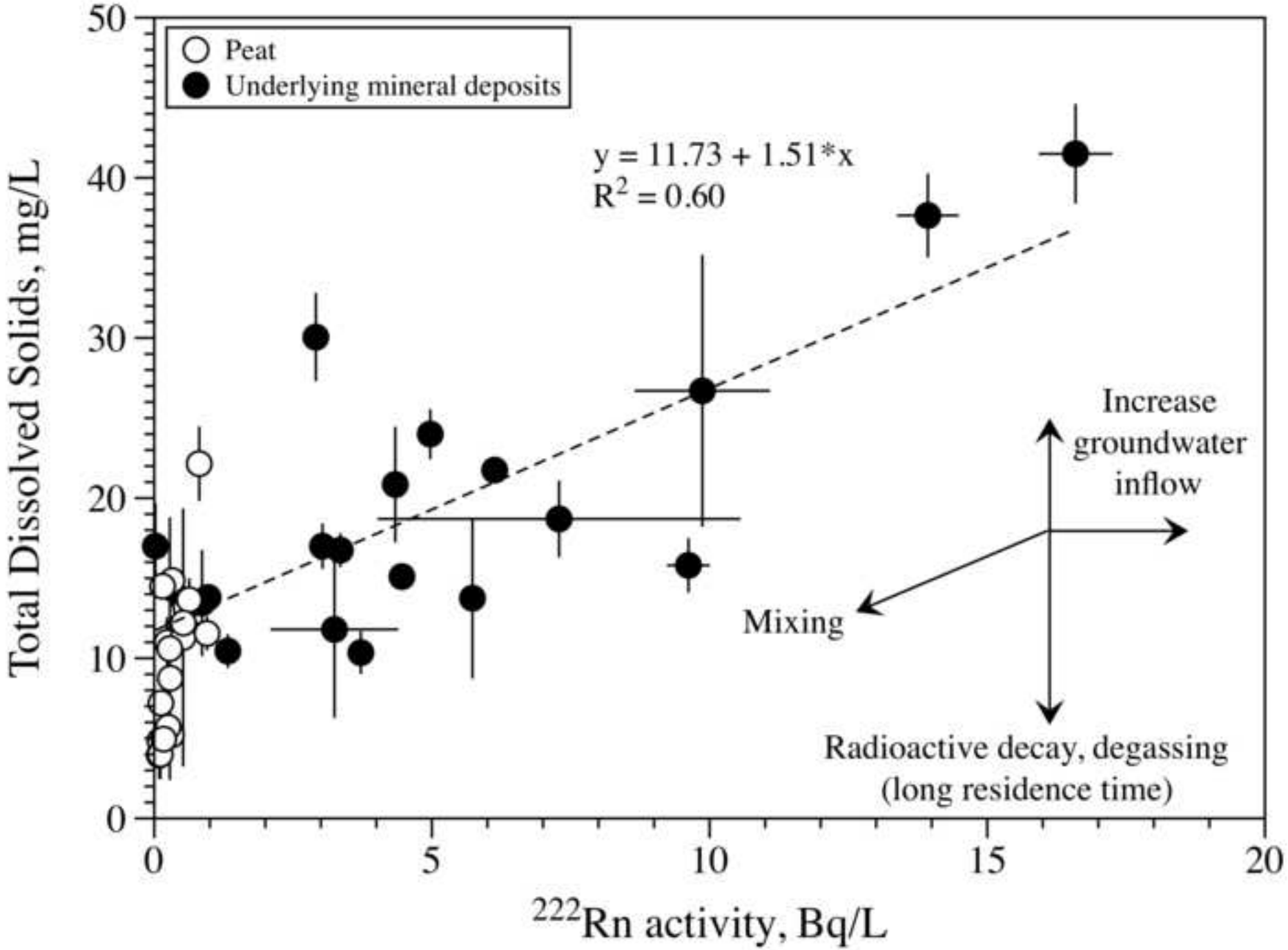

Figure 4

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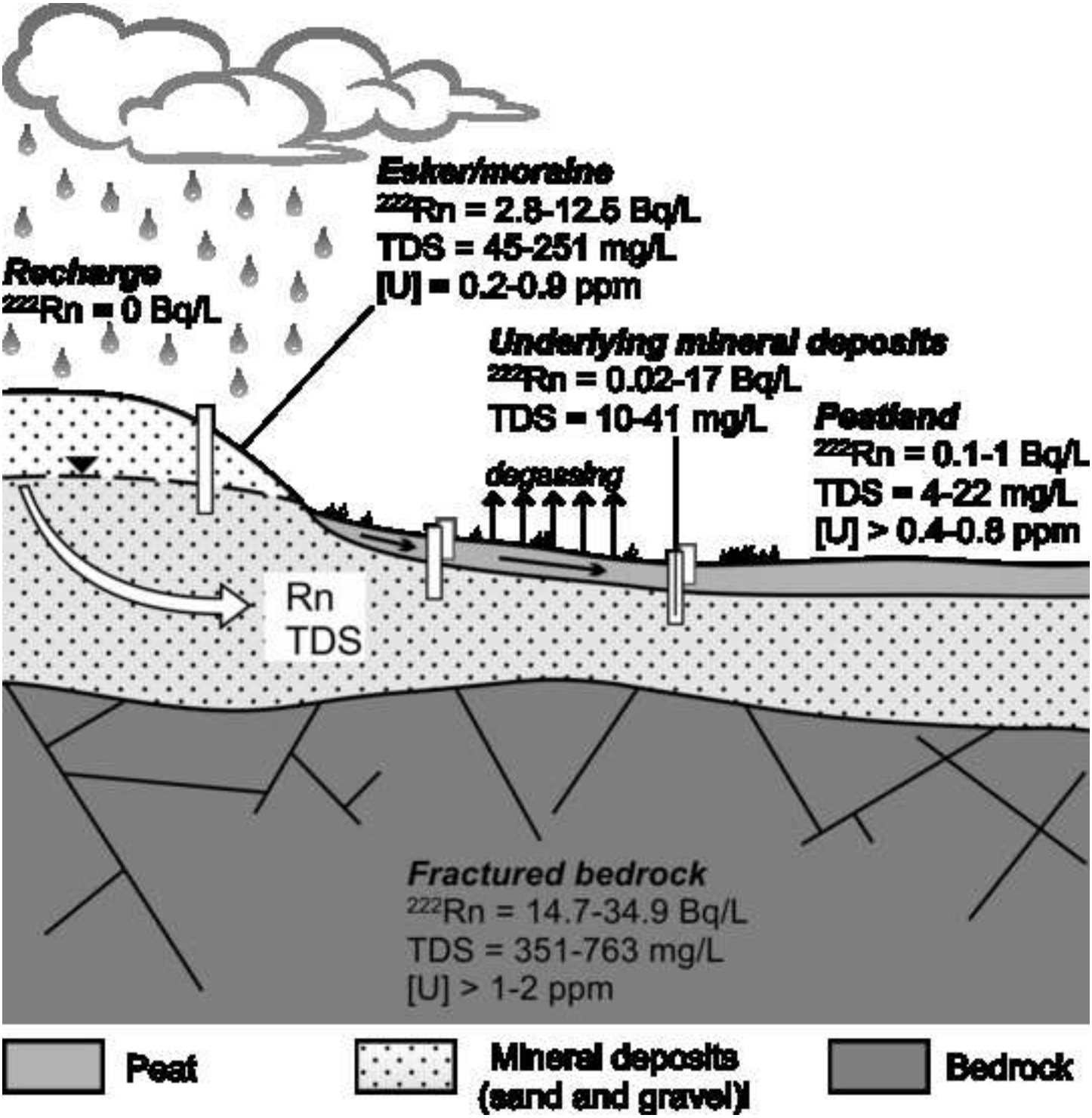

Table 1. Aquifer type, well depth and altitude, pH, TDS and ^{222}Rn activity from groundwater samples.

	Locality	Aquifer type	Altitude (m a.s.l.)	Well depth (m)	Screen depth* z_0 (m)	pH	TDS** mg/L	^{222}Rn Bq/L
Amos Municipality	SMB esker	Granular	312.3	23.3	23.2	8.3	106	11.1/11.5
PACES 1-12	SMB esker	Granular	337.9	53.5	52.8	8.6	103	5.9
PACES 1-13	SMB esker	Granular	337.9	22.9	22.2	8.9	86	2.8
TSAM-P1	SMB esker	Granular	310.0	10.5	9.8	8.5	92	4.9
TSAM-P2	SMB esker	Granular	310.6	16.6	15.9	8.3	128	7.9
TSSM-P1	SMB esker	Granular	314.6	26.8	26.1	8.7	92	4.3
TSSM-P3	SMB esker	Granular	309.1	27.6	26.9	8.9	96	9.9
TSSM-P5	SMB esker	Granular	331.8	29.0	27.5	7.5	96	3.5
Barraute Municipality	Barraute esker	Granular	305.0	22.9	20.6	7.9	251	9.4
Mont Video	Harricana Moraine	Granular	400.0	16.8	16.8	-	45	11.3
MDDEP S	Harricana Moraine	Granular	375.1	41.8	41.1	8.3	226	5.5
MDDEP P	Harricana Moraine	Granular	375.3	70.6	69.9	8.1	185	11.0
PACES 1-03	Harricana Moraine	Granular	375.0	64.4	63.7	8.4	115	12.5
Landrienne Municipality	Clay plain	Fractured	318.0	89.6	52.4	7.1	351	34.9
55HB	Clay plain	Fractured	316.0	109.8	62.2	7.2	763	15.0
101CH	Clay plain	Fractured	302.0	35.1	26.7	7.8	527	14.7

Error on ^{222}Rn activity is 4% and blanks 0.12 Bq/L (Lefebvre et al., 2013)

* For wells with complete casing, depth is taken from the surface to the half-lenght of the screen (4.5 and 1.5 m long)

* For open wells to bedrock, depth is taken from the surface to midway of the unscreened section.

** Total Dissolved Solids from Boucher et al. (2015)

SMB: Saint-Mathieu-Berry

Table 2

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Table 2. Piezometer depth, TDS, and ^{222}Rn activities

Piezometer name	Piezometer location	Distance from peatland edge (m)	Piezo depth (m)	TDS*** May mg/L	TDS*** Sept mg/L	^{222}Rn activity Bq/L
LB1.no4(p)	peat	226	1.1	4.0	5.7	0.1
LB1.no5(p)	peat	280	1.1	2.9	5	0.1
LB1.no6(p)	peat	460	1.1	2.9	5.1	0.1
LB1.no3(m)	mineral	172	1.6	18.9	15.1	0.02
LB1.no4(m)*	mineral	226	4.6	11.2	9.7	1.3/1.4
LB1.no5(m)	mineral	280	4.6	15.8	11.1	0.9
LB2.no5(p)	peat	150	1.1	7.5	6.9	0.1
LB2.no6(p)*	peat	353	1.1	4.7	5.7	0.4/0.2
LB2.no2(m)	mineral	11	1.6	18.0	16	3.0
LB2.no3(m)*	mineral	46	2.2	17.0	20.4	5.0/9.6
LB2.no5(m)	mineral	150	3.5	13.3	14.3	1.0
LC.no1*	esker	0	1.6	10.0	12	8.1/8.8
LC.no2(p)*	peat	34	0.4	5.7	n.d.	0.3
LC.no2(m)*	mineral	34	0.7	7.9	15.7	2.4/4.1
LC.no3(m)	mineral	59	1.2	19.0	15.1	4.5
LC.no4(m)*/**	mineral	109	0.8	12.5	14.8	0.3/0.8
LC.no6(m)*	mineral	368	3.3	32.7	20.7	9.0/10.7
SS.no2(p)	peat	41	1.1	14.9	14.8	0.3
SS.no3(p)	peat	160	1.1	14.0	12.3	0.6
SS.no4(p)	peat	211	1.1	12.1	9.9	0.2
SS.no5(p)	peat	256	1.1	8.8	8.7	0.3
SS.no6(p)	peat	473	1.1	12.7	14.6	0.6
SS.no2(m)	mineral	41	2.2	23.4	18.3	4.3
SS.no3(m)	mineral	160	4.0	39.5	35.8	13.9
SS.no4(m)*	mineral	211	3.9	43.7	39.3	6.9/16.6
SS.no5(m)	mineral	256	4.0	25.1	22.9	5.0
SS.no6(m)*	mineral	473	4.8	152.5	136.5	0.9/1.9
SMB.no4(p)*	peat	176	1.1	11.5	12.9	0.3/0.8
SMB.no5(p)	peat	228	n.d.	23.8	20.5	0.8
SMB.no6(p)	peat	431	0.9	10.8	12.3	1.0
SMB.no6(m)*	mineral	431	1.5	14.6	17	1.5/9.6

SN.no2(p)	peat	80	1.1	13.9	15.1	0.2
SN.no3(p)	peat	169	1.1	17.0	5.6	0.5
SN.no5(p)	peat	279	1.1	16.4	4.8	0.3
SN.no6(p)	peat	498	1.1	5.7	4.2	0.2
SN.no2(m)	mineral	80	2.3	32.0	28.1	2.9
SN.no3(m)	mineral	169	2.2	10.2	17.3	5.7
SN.no4(m)	mineral	225	3.0	22.1	21.4	6.1
SN.no5(m)	mineral	279	2.9	16.0	17.5	3.4
SN.no6(m)	mineral	498	3.3	9.4	11.3	3.7

*: indicates a piezometer that was sampled twice for radon activity

**: this sample had gas bubbles

***: TDS concentrations from Ferlatte (2014)

***Response to Reviewers**

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REVIEWER #1

(1) Clearly, there are quite a few special terminologies used in peatland research. I suggest the authors to revise the first and second paragraphs in the intro to define these terms, such as bogs, slope bogs, fens, eskers/moraines and underlying minerals clearly. I'm still unsure if the mineral layer underlying a peatland is part of an esker, and if bogs and slope bogs are the same. You also used slope bogs and slope peatlands interchangeably. If they are the same, why do you have to use slope bogs on Line 61 without a definition. All the terms throughout the manuscript should be used consistently and also must be defined when they first appear. All the elements important for understanding this study should be indicated in a diagram such as Figure 1.

We agree with the reviewer that these terms should be defined clearly. We believe that the first paragraph describes sufficiently eskers and moraines. In paragraph two, we have clarified the definitions of bog, fen, and slope bog. We have added a description of underlying mineral deposits in the Methods section.
We acknowledge that using interchangeably slope bog and slope peatland led to some confusion. We have introduced a sentence in paragraph two to explain that the system formed by a slope bog and a marginal fen is named slope peatland. We are now using this term throughout the manuscript.
We also added “underlying mineral deposits” and “esker” on Figure 1 for the reader to locate them clearly. The terms “fen” and “bog” were already included on the figure.

(2) On Page 12, 2 hypotheses were proposed to explain why second radon activity is generally higher than the first when two samples are taken consecutively from the same bore. However, these hypotheses were subsequently ruled out by analysing additional data. It is hard to believe that there is more than a factor of 2 difference in radon activity between 2 consecutive samples. It could be due to the problem of the field method. This matter needs to be further discussed and solutions to resolve this matter should be proposed in the discussion under a new section (which could be named as Limitations and improvement).

The additional data (i.e. noble gases) do not rule out our hypothesis but they confirm it. We better explained it at lines 283-285 of the new revised version. Indeed, the fact that the third purged volume (when the amount of water is sufficient) is un-degassed confirms that we need larger volume of water to be purged from the system to avoid to collect degassed samples. However, we agree with reviewer that a new section indicating limits of the methods and suggesting improvements has been added to the new revised version (4.3).

(3) Section "222Rn-depth gradients: an indicator of water mixing" reported two radon-depth gradients. I do not think you can derive the gradients. First, the three samples in the fractured bedrock were taken from 3 wells with long screens. They are not representative of the actual radon activities at the depths. There could be very significant mixing between different sources of groundwater. Hence, they are not supposed to be included to derive any relationships. They will result in misleading results as shown in the manuscript. Second, even if you include them in the figure, I do not think there are sufficient samples for you to derive such gradients. The Landrienne could simply be an outlier. If this is removed, the radon activities range between 5 and 25 Bq/L, which can be easily explained by radon variability. Unless more data are included, I do not agree with this section.

We agree that the relationship is tenuous and that does not bring additional data on the main topic of the manuscript, i.e. the groundwater inflow from eskers to peatlands. We thus decided to remove completely this section.

(4) In the last section in the discussion, lateral groundwater flow was discussed. I still do not understand how you can infer lateral groundwater flow. I understand lateral flow should occur in an ordinary hydrogeological setting as shown in Figure 5. You indicated in this section that vertical upward flow also occurs. So if groundwater with high

radon activity enters a peatland, the mixture of groundwater and peatland water will have very low radon. This can also explain why radon is low. I think this section needs to be revised carefully.

Yes indeed we rewrote this confusing section by clearly discussing on the TDS-relation with radon and what it brings of information on this study.

(5) My last comment relates to piezometers. I would like to see a figure (with 5 sub-figures) that shows hydraulic heads at all monitoring bores in all transects, each transect per sub-figure. I wonder if hydraulic gradients between two monitoring bores close to the marginal fens could help us infer lateral groundwater flow and verify radon results. Likewise, I wonder if nested bores could indicate vertical hydraulic gradients to determine vertical groundwater flow.

The reviewer is correct, hydraulic gradients between the monitoring bores close to the marginal fens are useful to infer lateral groundwater flow, and nested bores provide useful information to determine vertical groundwater flow. This data has been used in a previous study and recently published papers present figures with the detailed information the reviewer is asking for (we do not think it is necessary to reproduce these figures here). In their Figure 4, Ferlatte et al. (2015) present the heads (average, minimum and maximum values) measured in the peat deposits (upper piezometer) and in the underlying mineral deposits (lower piezometer). In their Figure 5, these authors present a typology for lateral flow connections in slope and basin peatlands. Ferlatte et al. (2015) also include a complete discussion on vertical groundwater flow which is summarized in their Table 3 (dominating flows are downwards with some exceptions of vertical inflows). To make previous knowledge about the sites clearer in the manuscript, we have included additional information on Ferlatte et al (2015) towards the end of the Study area section.

Some minor comments:

(1) Radon expression needs to be consistent throughout the paper. Use 222Rn or Radon.

We believe that 222Rn or radon can be used interchangeably without creating any confusion, but we mainly changed to 222Rn to be consistent throughout the text.

(2) On line 219, explain why using 20-50 cm3 of scintillant instead of a constant volume.

Sorry, depending from the size of used sampled bottles. In this case, 25 cm^3 of scintillant was constantly used.

(3) On line 272, as mentioned above, please distinguish between eskers and mineral deposits. Figure 5 used the same texture for the esker and the mineral, which makes me more confused.

Mineral deposits and peatlands have not the same texture in figure 5, now figure 4 (mineral deposits are in dotted texture while peat is grey). We corrected the figure and the legend that was confusing.

(4) On lines 302-334, the comparison is confusing. First, where are Fennoscandian regions? Are they close by with a similar setting? If yes, then it is reasonable to say high radon activities are expected. Otherwise, whatever observed is reasonable. Second, I think you probably do not need to show in the manuscript that you did back-of-envelope calculation to prove your measured radon activities are correct. It does not offer any help with understanding. Rather, it is distracting.

Yes Fennoscandian region has a very similar geological environment than Abitibi: an Archean bedrock with directly on Quaternary age eskers and moraines produced during the last glaciation. Lithologies and glaciated landform are very similar to those of Northern Canada. Yes we agree that back-of-calculation are distracting so we reduced the explanation to 6 lines (317-322 in the revied manuscript).

(5) On lines 402 and 404, one median in the peatland is 0.9 Bq/L but the other is 0.3 Bq/L. This needs clarification.

There was confusion between median and average. All data reported are average values and corrected in both text and figures.

(6) Lines 454-467 are more about future work instead of conclusions. Move this paragraph to the discussion (e.g., under the section I proposed) and summarise it with 1-2 sentences in the conclusion.

As above mentioned we wrote a new section 4.3 describing limits and improvements of the method.

Reviewer #2

This paper explores the potential for 222Rn to trace groundwater flows into peatlands in the Amos region of Canada.

The paper is very well written, clear, concise, and I feel needs little work to bring it to publication standard.

I only have a few minor comments.

I think the Study area section could be reduced a bit, for example combining much of the technical data (e.g. water ages etc) into a table.

Geological description was reduced but adding water ages (that are already published in Boucher et al. (2015)) does not help in shortening the section.

Methods, explain how the decay correction was applied (Line 209)

This is a classic equation and we think is superfluous to report it.

Results, Break into smaller paragraphs, (for example Line 271 to Line 298 is 1 paragraph). This could be achieved by removing the more "discussion" type sections (line 280 to 291) to the discussion.

Yes, the results section has been modified and improved.

Line 409 .... degasses and decays?

Yes, this section has been largely rewritten showing the effects of different processes (including decay and degassing).

REVIEWER #3

In this study, radon was tested as a potential groundwater tracer in peatland ecosystems where these groundwater interactions are not well understood and require further investigation. This highlights the novel technique of using radon to trace groundwater inflows into peatlands, building on previous work in the region that mainly utilised TDS to explain groundwater flows. The methods used are thoroughly explained and easy to follow while the entire manuscript is very well written. The hypotheses are well outlined, tested and explained. The discussion relates the results well with other literature and gives good explanations of groundwater inflow derived from radon observations. There is a good section describing the possibility of using radon as a quantitative tracer in the future and improving the methods for radon sampling. Considering this is the first time radon has been sampled from peat water, this is an excellent and invaluable investigation, and I recommend it for publication.

A few general comments

Using TDS values from 2010 is okay and it is good that samples were taken in both May and September with little difference between the two months. However, sampling for these parameters at the same time as radon would have been preferable as conditions can change. I would suggest that future radon sampling be done in conjunction with major ion sampling.

Yes, we agree.

It is usual to purge 3 well volumes prior to groundwater sampling to ensure the sample is from the aquifer. I understand this was not possible in this situation where recharge into the piezometer is too slow. This may have produced a radon value less than the actual amount from the peatlands. It is good that possible methodological improvements are suggested for radon sampling and it would be an interesting further study to conduct.

Please note that this has been done for aquifers. Municipal wells are in continuous flow, so after a few minutes, parameters (T, pH, electric conductivity etc.) are stabilized and water can be taken. Water from domestic wells are also taken after parameters stabilized (as indicated in the text) and this often after a volume of water that is well beyond the 3 volumes minimal required. Water from peatland was purged only the equivalent of 1 well volume because of the limited amount of water available. We added a new section 4.3 where limits and improvements of the method are described.

A few specific comments

Line 31 – "222Rn activity was measured in groundwater from 16 piezometers, municipal and domestic wells from the Saint-Mathieu-Berry and Barraute eskers, from the Harricana Moraine, and from the fractured bedrock."

This is initially a confusing sentence because I was not sure if the number 16 referred only to the piezometers or also to the wells. It might be better to state the number of samples taken from the eskers, moraines and bedrock instead. To be clearer on the number of samples taken, you can change the sentence to read "222Rn activity was measured in groundwater from piezometers, municipal and domestic wells from the Saint-Mathieu-Berry and Barraute eskers (n=9), from the Harricana Moraine (n=4), and from the fractured bedrock (n=3)."

The sentence has been changed. Also we put order in the "Method" section because the newly drilled and instrumented piezometer from UQAT are sometimes designated as wells, sometimes as piezometers. This has been corrected.

Line 387 – It appears in Figure 3 that some wells in the moraines are deeper than the wells in the fractured bedrock. However, you have mentioned that the moraines are shallower than fractured bedrock and this is the foundation of your radon vs depth relationship, where radon is higher in the deeper fractured bedrock than the shallower moraines and eskers. For the relationship on the right in Figure 3, your second hypothesis makes sense but the relationship on the left has moraines wells that are deeper than the fractured bedrock wells. The second hypothesis can only explain the relationship on the right. Further explanation is needed for the relationship on the left because if looking only at Figure 3, this does not appear correct. I understand the fractured bedrock is beneath the moraines and eskers. Instead of referring solely to depth, state the fractured bedrock as underlying the moraines and eskers,

rather than being deeper because the nature of the terrain and the distance between sites does not necessarily mean that the fractured bedrock wells will be deeper than the moraines and eskers.

This section was removed as suggested by reviewer no. 1 because of the poor constraints. If corrected for altitude the depth of wells (altitude is now reported in Table 1) the relation is blurred and lose significance.

Line 398 – You mentioned earlier that advection was not a possible explanation for radon and depth gradients because advection velocity would have to be 65 times faster the radon-bearing advection air velocity in soil. However, to explain the second hypothesis, you mention that radon advection must be rapid. Can you please explain this further to put it into context with your above advection rate comment and give a value to this concluded advection rate?

This section has been removed.

Line 428 – You mention local vertical groundwater inflow occurs on some occasions at a limited number of stations but don't give an explanation as to the mechanisms that create the vertical inflow rather than the downward vertical flow observed at most other stations. Could it be that these are sites that are more fractured compared to other sites? Why are the radon concentrations higher here? Establishing horizontal inflows had already been down with TDS so the fact that you've found radon hotspots that indicate vertical upwards flow is novel and warrants more explanation. Also, what were the TDS values at these radon hotspot sites?

All this section has been simplified and the TDS-Radon relation better explained.

Revision with Track Changes

Click here to download Revision with Track Changes: JENVRAD-D-16-00246_Text_Modif.docx