



# Utility of $^{222}\text{Rn}$ as a passive tracer of subglacial distributed system drainage



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## ABSTRACT

Water flow beneath the Greenland Ice Sheet (GrIS) has been shown to include slow-inefficient (distributed) and fast-efficient (channelized) drainage systems, in response to meltwater delivery to the bed via both moulins and surface lake drainage. This partitioning between channelized and distributed drainage systems is difficult to quantify yet it plays an important role in bulk meltwater chemistry and glacial velocity, and thus subglacial erosion. Radon-222, which is continuously produced via the decay of  $^{226}\text{Ra}$ , accumulates in meltwater that has interacted with rock and sediment. Hence, elevated concentrations of  $^{222}\text{Rn}$  should be indicative of meltwater that has flowed through a distributed drainage system network. In the spring and summer of 2011 and 2012, we made hourly  $^{222}\text{Rn}$  measurements in the proglacial river of a large outlet glacier of the GrIS (Leverett Glacier, SW Greenland). Radon-222 activities were highest in the early melt season (10–15 dpmL<sup>-1</sup>), decreasing by a factor of 2–5 (3–5 dpmL<sup>-1</sup>) following the onset of widespread surface melt. Using a  $^{222}\text{Rn}$  mass balance model, we estimate that, on average, greater than 90% of the river  $^{222}\text{Rn}$  was sourced from distributed system meltwater. The distributed system  $^{222}\text{Rn}$  flux varied on diurnal, weekly, and seasonal time scales with highest fluxes generally occurring on the falling limb of the hydrograph and during expansion of the channelized drainage system. Using laboratory based estimates of distributed system  $^{222}\text{Rn}$ , the distributed system water flux generally ranged between 1–5% of the total proglacial river discharge for both seasons. This study provides a promising new method for hydrograph separation in glacial watersheds and for estimating the timing and magnitude of distributed system fluxes expelled at ice sheet margins.

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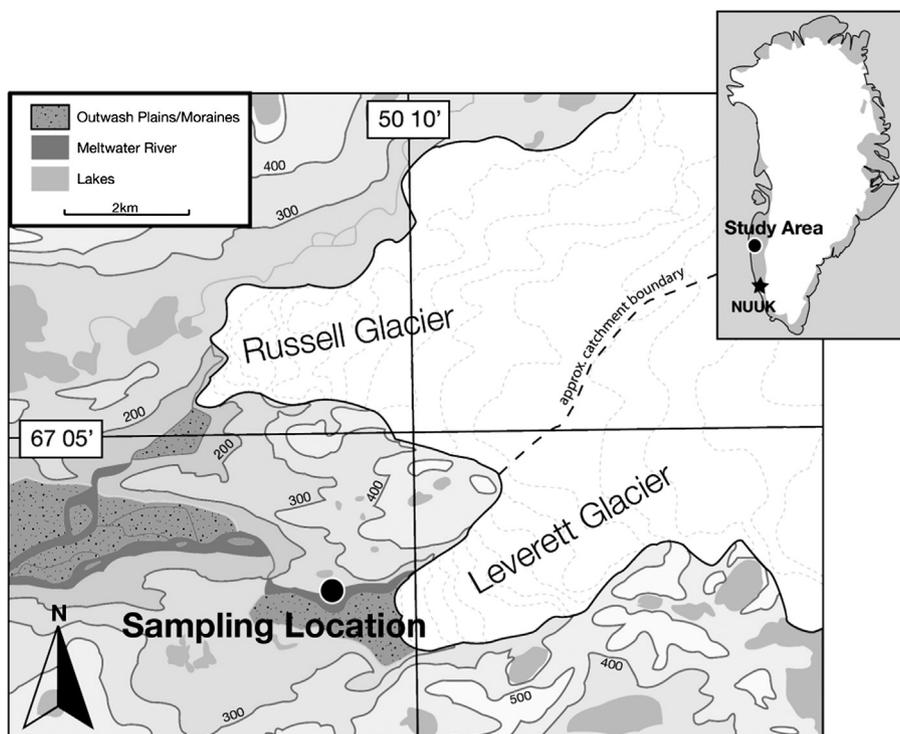
## 1. Introduction

Beneath the ablation zone of the Greenland Ice Sheet (GrIS), meltwater flow paths influence glacier velocities and bulk meltwater chemistry (Bartholomew et al., 2011a, 2012; Tedstone et al., 2013; Hawkings et al., 2014). After the onset of spring melt, the majority of surface derived meltwater travels to the glacier or ice sheet bed through fractures, crevasses and moulins (Sharp et al., 1993; Das et al., 2008). During the summer, bulk meltwater is largely composed of two components, channelized drainage and

distributed drainage, which are both derived from snow and ice melt (Tranter, 1993; Collins, 1979; Chandler et al., 2013; Cowton et al., 2013). On the time scale of an entire melt season, the major component is channelized flow, which pertains to meltwater moving efficiently through large basal channels (Röthlisberger, 1972; Nye, 1973). In contrast, distributed drainage refers to meltwater in slow transit through either cavities that open behind bedrock bumps due to ice sliding (linked-cavity system; Walder, 1986), a water sheet of near uniform thickness at the ice bed (Creys and Schoof, 2009), or water flow through permeable subglacial till (Boulton et al., 2009). Meltwater traveling through distributed systems influences subglacial hydraulic pressure distribution, channel spacing, basal sliding and bed deformation (Boulton et al., 2009; Rempel, 2009). In general, meltwater in the distributed system tends to have elevated dissolved solid concentrations, a feature

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**Fig. 1.** Location of Leverett Glacier in west Greenland. The primary sampling location for continuous  $^{222}\text{Rn}$  measurements is indicated by the black circle though some early season deployments of the  $^{222}\text{Rn}$  sensor occurred much closer to the glacier terminus. Figure adapted from [Hawkings et al. \(2014\)](#).

exploited by early attempts at hydrograph separation in glacial watersheds ([Collins, 1979](#)). However, rapid mineral weathering reactions may occur when sediments traveling through closed,  $\text{CO}_2$  limited, distributed systems mix with open-system, low ionic strength channelized meltwater ([Raiswell, 1984](#); [Tranter, 1993](#); [Sharp et al., 1993](#)). Consequently, solute concentrations, often inferred through electrical conductivity (EC) measurements, cannot unequivocally be used as conservative tracers of distributed system meltwaters.

Radon-222 has been used extensively to examine groundwater-surface water exchange processes in a wide range of freshwater and marine systems; this is because groundwater  $^{222}\text{Rn}$  activities are typically highly enriched relative to surface waters due to radioactive decay of  $^{226}\text{Ra}$ , naturally present in aquifer mineral surfaces ([Burnett and Dulaiova, 2003](#); [Cook et al., 2003](#); [Dulaiova et al., 2008](#); [McCallum et al., 2012](#)). As a relatively soluble, inert noble gas that does not participate in biogeochemical or weathering reactions once in solution,  $^{222}\text{Rn}$  ( $t_{1/2} = 3.82$  days) can only be added to glacial meltwater in measurable quantities from extensive water-rock interactions. Hence, by its nature, distributed system meltwater should acquire significantly higher  $^{222}\text{Rn}$  activities than meltwater that flows through open channels.

Discrete  $^{222}\text{Rn}$  measurements in small glacial watersheds have been used to infer the transition from distributed to channelized drainage ([Kies et al., 2011](#); [Bhatia et al., 2011](#); [Kies et al., 2015](#)). This paper expands upon these earlier studies by examining the utility of long term continuous  $^{222}\text{Rn}$  measurements in the proglacial river of a large GrIS outlet glacier during the spring and summer of 2011 and 2012. Using a detailed mass balance modeling approach, we provide evidence that  $^{222}\text{Rn}$  is a passive tracer of subglacial hydrological routing that can be used to infer the timing and magnitude of distributed system fluxes. To our knowledge this is the first time that continuous, high-resolution  $^{222}\text{Rn}$  measurements have been reported from a proglacial river over the course of a melt season.

## 2. Methods

### 2.1. Study area

Fieldwork was conducted during the 2011 and 2012 melt seasons at Leverett Glacier, a large land-terminating glacier on the western margin of the GrIS ( $67^{\circ}03'57.81''\text{N}$ ,  $50^{\circ}10'01.83''$ ; [Fig. 1](#)). Its hydrological catchment covers  $>600$  km<sup>2</sup>, reaches an elevation of 1500 m, and ranges in width from 10–40 km ([Bartholomew et al., 2011a, 2011b](#)). Meltwater from the catchment is channelized through a single large proglacial river (Leverett River); at peak discharge, typical flows from the river are in the range of  $300\text{--}400$  m<sup>3</sup> s<sup>-1</sup> ([Bartholomew et al., 2011a, 2011b](#); [Cowton et al., 2012](#)). During an exceptional melting period in 2012, the river reached  $\sim 800$  m<sup>3</sup> s<sup>-1</sup> ([Tedstone et al., 2013](#)). On the south side of Leverett Glacier's snout, the bedrock is a late Achaean (2.5 Ga) granite ([Escher, 1971](#); [Nutman et al., 2010](#)) while the Ikertoq complex (1.85 Ga), composed of basement gneisses and granites, borders the north side ([Henriksen et al., 2009](#)). The proglacial valley is filled with quaternary sediments composed of weathered materials from afore mentioned rock units ([Hindshaw et al., 2014](#)). This suggests that the subglacial lithology is similar to the bedrock adjacent to the glacier's snout.

### 2.2. Discharge, conductivity, suspended sediment, and $^{226}\text{Ra}$ measurements

Proglacial river discharge was measured using continuous water stage monitoring through a stable bedrock section of the river ([Sole et al., 2013](#); [Tedstone et al., 2013](#)). Stage was converted to discharge using a continuous stage-discharge curve created from repeat Rhodamine WT and Rhodamine B dye injections throughout both melt seasons; the normalized root mean squared deviation of the discharge record has been estimated to be  $\pm 10\%$  ([Tedstone et al., 2013](#)). Concurrent measurements of EC and the suspended sediment concentration (SSC) are also presented here and, for 2011

and 2012 data, in Butler (2014) and Hawkings et al. (2014) respectively. EC was recorded every five minutes using a Campbell Scientific 247 combined temperature-EC sensor, and logged using Campbell Scientific CR1000 and CR800 loggers. EC was calibrated using a KCl solution of known concentration; errors on EC measurements were  $\pm 10\%$  (1-sigma). SSC was estimated from turbidity measurements made with a Partech IR 15C turbidity probe. Calibration curves were created from discrete suspended sediment samples collected from the river. Errors on SSC were  $\pm 6\%$  (1-sigma). Dissolved  $^{226}\text{Ra}$  was measured throughout the 2011 and 2012 field seasons using methods described by Charette et al. (2001). Briefly,  $\sim 200$  L river samples were filtered through a column packed with  $\text{MnO}_2$  impregnated fiber. The fiber was ashed, packed in a sealed plastic vial, and counted for 2–3 days on a well-type germanium gamma detector (Canberra) calibrated using a NIST  $^{226}\text{Ra}$  standard prepared in the same manner as the samples.

### 2.3. Measuring radon in the proglacial river

Radon-222 was measured in the Leverett Glacier proglacial river during 2011 (May 8–August 5) and 2012 (May 12–July 28). Continuous (hourly) measurements were made using a RAD7 (DurrIDGE Inc.) radon-in-air monitor in series with a desiccant chamber and a RAD7 water probe, a submersible air-filled gas-permeable membrane coil made from Accruel® tubing (henceforth the term “water probe” will refer to the  $^{222}\text{Rn}$  extraction unit). River water  $^{222}\text{Rn}$  equilibrates with air in the membrane coil through passive diffusion; the air is continuously circulated in a closed loop through the RAD7 radon-in-air monitor system (Hofmann et al., 2011; Schubert et al., 2012). Radon-in-air activities were then converted to radon-in-water activity via the temperature dependent air-water partition coefficient as described by Schubert et al. (2012). The water probe was deployed on the northern bank of the river as close to the glacier’s terminus as possible (0–1 km) to minimize potential evasion of  $^{222}\text{Rn}$  from the river to the atmosphere (Supplementary Material). Rising river stage required that we periodically moved the water probe downstream to a more stable riverbank. There was no observed decrease in effectiveness of the water probe during long term deployment (Supplementary Material). Typical 1-sigma counting errors for the water probe  $^{222}\text{Rn}$  activities were  $\pm 5$ –20%. Discrete  $^{222}\text{Rn}$  samples were collected upstream of the water probe at the ice sheet terminus in 250 mL bottles and analyzed using a RAD7 plus Rad-H<sub>2</sub>O attachment on a regular basis (Fig. S1; Supplementary Material). These discrete  $^{222}\text{Rn}$  samples agree within the counting errors of the corresponding continuous  $^{222}\text{Rn}$  measurements (Fig. S1; Supplementary Material).

Measurements of  $^{222}\text{Rn}$  via the water probe system may lag actual aqueous  $^{222}\text{Rn}$  due to the time required to equilibrate  $^{222}\text{Rn}$  across the gas-permeable membrane (Schubert et al., 2012). To assess the magnitude of this offset, we analyzed field equilibration times during instrument startup and performed laboratory experiments to estimate the water probe’s response time to changes in  $^{222}\text{Rn}$  activity. Key factors that control the equilibration time include the air volume in the system (RAD7 unit, tubing, desiccant and membrane coil), the membrane coil surface area, and the  $^{222}\text{Rn}$  activity gradient across the membrane water/air interface (Schubert et al., 2012). The equilibration time can be minimized when the air volume of the system is low (i.e. using less tubing between RAD7 and membrane), the membrane interface area is maximized (i.e. lengthening the membrane tubing), and the water/air  $^{222}\text{Rn}$  activity gradient across the membrane coil is large. The  $^{222}\text{Rn}$  activity gradient at the membrane water/air interface has by far the largest effect on the equilibration and response time of the water probe; in stagnant water the response time is on the order of several hours (Hofmann et al., 2011;

Schubert et al., 2012). In a flowing river, the activity gradient at the water/air interface is close to 100% such that the response time can be reduced to  $< 1$  h, similar to results obtained using a spray chamber equilibrator such as the RAD-Aqua (DurrIDGE Inc.; Schubert et al., 2012). In our case, the equilibration time of the water probe in the proglacial river was determined to be no greater than 2 hours based on laboratory experiments and less than 1 hour in field observations. A detailed description of these tests and associated results is presented in the Supplementary Material.

### 2.4. $^{222}\text{Rn}$ in subglacial distributed systems and sediment properties

Because we were unable to obtain *in-situ* distributed system samples from beneath the ice sheet, we estimated  $^{222}\text{Rn}$  in this environment via laboratory-based equilibration experiments (Corbett et al., 1998; Dulaiova et al., 2008) using sediments discharged from the subglacial environment. For these tests, four separate aliquots of sediment (50 g) from the proglacial river were incubated with Ra-free water in a sealed 1 L high-density polyethylene bottle for greater than five half-lives ( $> 20$  days). Samples were flushed into a cold trap and scintillation cells using helium and analyzed in triplicate on alpha scintillation counters (Corbett et al., 1998). Wet sediment  $^{222}\text{Rn}$  activities in  $\text{dpm g}^{-1}$  were converted to pore water  $^{222}\text{Rn}$  activities ( $\text{dpm L}^{-1}$ ) using wet bulk densities and porosities (Supplementary Material).

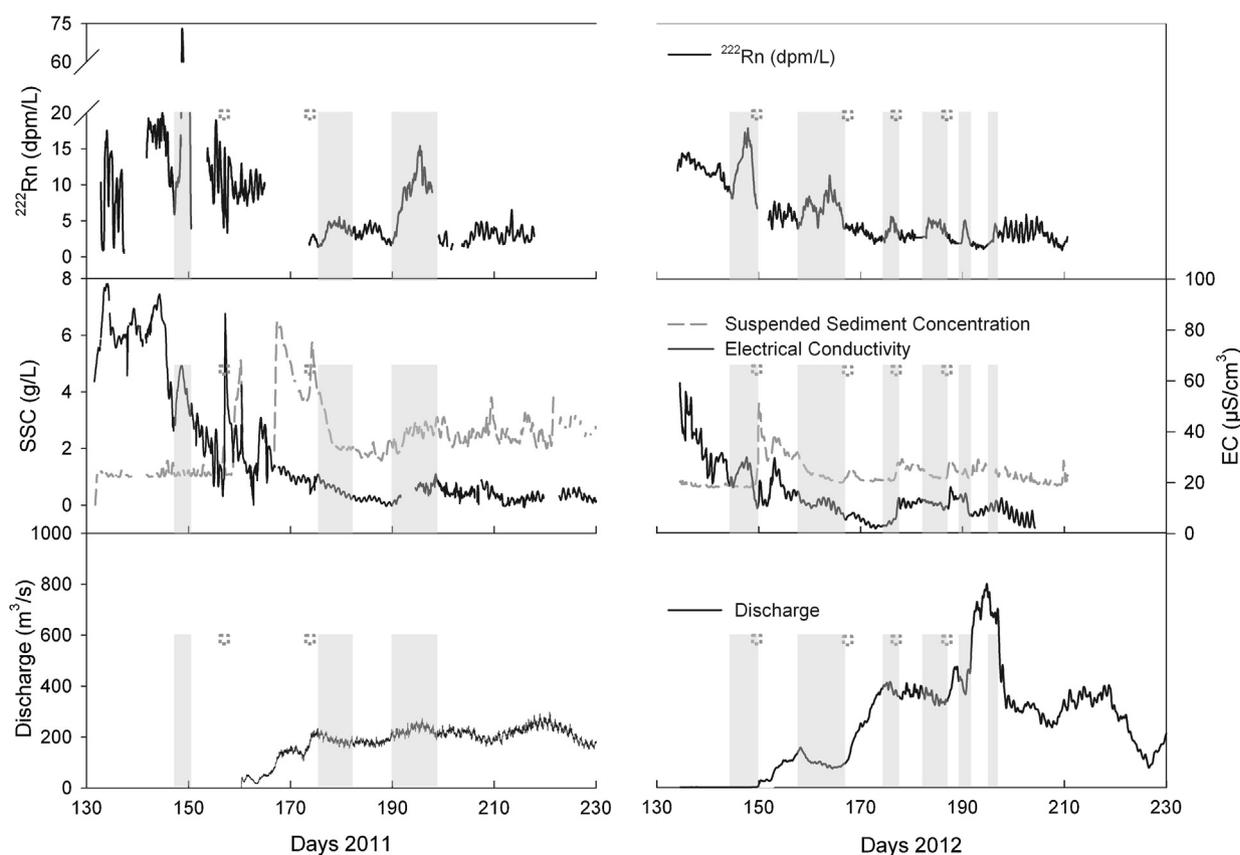
Radon-222 diffusion from sediments in subglacial channels was another potential source of  $^{222}\text{Rn}$  beneath the GrIS. We employed a laboratory method described by Chanyotha et al. (2014) to quantify the diffusive flux. Briefly, 100 g of wet subglacial sediment and 500 mL of  $^{222}\text{Rn}$ -free water were sealed inside a gas tight reaction flask connected in a closed loop with a RAD7. Air was pumped through a gas diffusion stone immersed in the water phase using the built in RAD7 pump, then through the desiccant, and back to the radon analyzer where the activity was measured and recorded. While gas leakage is not an issue for routine measurements, a known small leak within the internal air pump of the RAD7 was corrected during the multi-day experiment according to the approach described in Chanyotha et al. (2014). The diffusive flux was determined from the near-linear slope of  $^{222}\text{Rn}$  activity in the reaction flask versus time over the first several hours of the experiment. Slope uncertainty was used to estimate the uncertainty of the diffusive flux.

Leakage is an issue for this approach, not necessarily from diffusion through the walls of the plastic bottle but from a known small leak in the internal air pump of the RAD7 as described in Chanyotha et al. (2014). This is not an issue for routine measurements with the RAD7, but needs to be corrected for in the multi-day sediment equilibration method. The data has been leak corrected according to the approach described in Chanyotha et al. (2014).

## 3. Results

### 3.1. Discharge

Warmer average air temperatures resulted in nearly twice as much annual discharge in 2012 as in 2011, corresponding to  $\sim 2.4$  and  $1.4 \text{ km}^3$ , respectively within the Leverett Glacier catchment (Fig. 2). Average flows of the proglacial river in the summer were  $\sim 200 \text{ m}^3 \text{ s}^{-1}$  in 2011 (Sole et al., 2013) and  $\sim 400 \text{ m}^3 \text{ s}^{-1}$  in 2012 (Tedstone et al., 2013). In 2012, the largest melting event since at least 1889, as indicated by ice cores at the Summit Station, occurred on July 12th (day 194) when over 98% of the surface of the GrIS experienced melting (Nghiem et al., 2012). During this period, river discharge reached  $\sim 800 \text{ m}^3 \text{ s}^{-1}$  (Tedstone et al., 2013), almost three times larger than maximum discharge in 2011 (Fig. 2;



**Fig. 2.** Results from 2011 and 2012 field seasons. Top panels: radon-222 activity ( $\text{dpm L}^{-1}$ ); middle panels: EC and SSC (Hawkings et al., 2014); bottom panels: river discharge (Sole et al., 2013; Tedstone et al., 2013). Shaded grey boxes represent  $^{222}\text{Rn}$  peaks while dashed grey boxes are SSC/EC peaks, which likely correspond to supraglacial lake drainage events (discussed in text).

Sole et al., 2013). Furthermore, in 2011 the river was largely ice-covered until day 160, while in 2012 the river was ice free from the start of sampling (day 133) onward.

### 3.2. EC and SSC

We used EC as a proxy for the concentration of dissolved solutes in the proglacial river. During both 2011 and 2012, EC was elevated in the early season ( $60\text{--}100 \mu\text{S cm}^{-1}$ ) and decreased to  $10\text{--}20 \mu\text{S cm}^{-1}$  with increasing river discharge (Fig. 2; Butler, 2014; Hawkings et al., 2014). These results are consistent with the EC range reported in 2009 (Bartholomew et al., 2011a, 2011b). Butler (2014) and Hawkings et al. (2014) observed diurnal variations in EC through much of the 2011 and 2012 seasons, which in general were inverse to river discharge. During both seasons, peaks in EC punctuated the record.

The SSC varied between  $1\text{--}7 \text{ g L}^{-1}$  in 2011 (Butler, 2014) and  $1\text{--}4 \text{ g L}^{-1}$  in 2012 (Hawkings et al., 2014); these concentrations were similar to values reported for 2009 and 2010 (Cowton et al., 2012). Like EC, diurnal variations in SSC occurred throughout much of the 2011 and 2012 melt seasons, however, these diurnal variations in SSC generally increased with rising river discharge. In 2012, peaks in SSC and EC generally occurred simultaneously (Fig. 2, dashed boxes) as noted by Hawkings et al. (2014) and in line with observations in 2009 (Bartholomew et al., 2010). In 2011, SSC and EC peaks were largely decoupled (Fig. 2).

### 3.3. Radon-222 in the Proglacial River

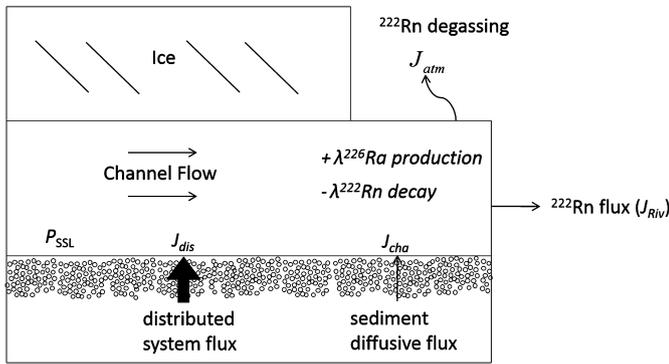
In both 2011 and 2012, the highest  $^{222}\text{Rn}$  activities were observed in the early season and generally decreased with increasing river discharge (Fig. 2). In 2011, typical  $^{222}\text{Rn}$  activities in the early

season (days 132–160) were between  $10\text{--}15 \text{ dpm L}^{-1}$ . Following day 170, activities were generally between  $2\text{--}5 \text{ dpm L}^{-1}$ . In 2012, higher  $^{222}\text{Rn}$  activities were observed in the early season, typically between  $5\text{--}18 \text{ dpm L}^{-1}$ , falling to between  $2\text{--}10 \text{ dpm L}^{-1}$  after day 150. In a small proglacial river  $\sim 100 \text{ km}$  north of our field site, Bhatia et al. (2011) reported significantly higher  $^{222}\text{Rn}$  activities ( $25\text{--}76 \text{ dpm L}^{-1}$ ), though peak river discharge rates ( $\sim 2 \text{ m}^3 \text{ s}^{-1}$ ) were orders of magnitude lower than at Leverett Glacier. Regardless, activities measured in this study and in Bhatia et al. (2011) are much higher than those generally observed in non-glaciated river catchments. For example, both Cook et al. (2003) and McCallum et al. (2012) report maximum  $^{222}\text{Rn}$  activities of  $\sim 1 \text{ dpm L}^{-1}$  in tropical and temperate rivers.

Significant peaks in  $^{222}\text{Rn}$  are highlighted in Fig. 2 by grey shaded boxes. In 2011,  $^{222}\text{Rn}$  peaked at  $\sim 75 \text{ dpm L}^{-1}$  in the early season (days 148–150); during this time, the river was ice covered and a small upwelling spring appeared at the glacier portal through which nearly all river discharge originated. A second large peak (days 190–200) was observed in  $^{222}\text{Rn}$  when activities climbed to  $\sim 15 \text{ dpm L}^{-1}$  from a pre-peak baseline of  $3 \text{ dpm L}^{-1}$  over a  $\sim 5$  day period (Fig. 2). In 2012,  $^{222}\text{Rn}$  peaks occurred regularly, approximately once every 8–10 days throughout the season (Fig. 2). Radon-222 peaks in 2012 generally increased on the falling limb or inflection point of the hydrograph and decreased when river discharge rose (Fig. 2). This general relationship between discharge and  $^{222}\text{Rn}$  was not observed in 2011. In both field seasons,  $^{222}\text{Rn}$  did not correlate with SSC or EC.

### 3.4. Radon activity in distributed system meltwater

Laboratory derived distributed system  $^{222}\text{Rn}$  activities ( $\text{Rn}_{\text{dis}}$ ) were estimated using the  $^{222}\text{Rn}$  activity of porewater in sediments



**Fig. 3.** Summary of  $^{222}\text{Rn}$  sources and sinks in the proglacial river. Sources of  $^{222}\text{Rn}$  include the distributed system flux ( $J_{dis}$ ),  $^{222}\text{Rn}$  diffusion through sediments into channels and cavities ( $J_{cha}$ ),  $^{226}\text{Ra}$  bound to the surface of the suspended sediment load ( $P_{SSL}$ ), and the decay of dissolved  $^{226}\text{Ra}$  ( $\lambda^{226}\text{Ra}$ ). The sinks of  $^{222}\text{Rn}$  include gas loss to the atmosphere ( $J_{atm}$ ) and radioactive decay of  $^{222}\text{Rn}$  ( $\lambda^{222}\text{Rn}$ ). The flux of  $^{222}\text{Rn}$  from the river ( $J_{riv}$ ) is the summation of these variables.

collected from the proglacial river (Section 2.4) and sediment properties (Corbett et al., 1998) including bulk density ( $\rho_B$ ) and porosity ( $\phi$ ; Supplementary Material). Wet sediment  $^{222}\text{Rn}$  activities ( $\text{Rn}_{sed}$ ) were 0.064–0.093 dpm g $^{-1}$  (avg = 0.076 dpm g $^{-1}$ ,  $n = 4$ ).  $\rho_B$  and  $\phi$  were identical for the three glacial river sediment samples analyzed and were 1.7 g cm $^{-3}$  and 0.37 respectively. Our results are consistent with those of Dow et al. (2013), who estimated subglacial sediment  $\phi$  in the Leverett Glacier catchment between 0.3–0.4. Using Eq. (1) and assuming that subglacial sediment  $\phi$  varied between 0.3–0.4, that  $\text{Rn}_{sed}$  was between 0.064–0.093 dpm g $^{-1}$ , and that  $\rho_B = 1.7$  g cm $^{-3}$ ,  $\text{Rn}_{dis}$  would be expected to span from 270–530 dpm L $^{-1}$ .

$$\text{Rn}_{dis} = \frac{\text{Rn}_{sed} \times \rho_B}{\phi} \times 1000 \text{ (g L}^{-1}\text{)} \quad (1)$$

These values are lower than laboratory derived pore water  $^{222}\text{Rn}$  activities in proglacial sediments reported by Bhatia et al. (2011), which were between 1285–3045 dpm L $^{-1}$ . A number of factors could explain the difference between the two sites: including sediment grain size (higher surface area/volume), degree of weathering (affects sediment  $^{226}\text{Ra}$  parent activities), or  $^{238}\text{U}$  content of the sediment (Dulaiova et al., 2008).

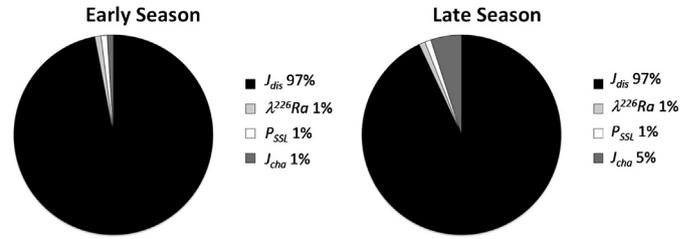
## 4. Discussion

### 4.1. Radon-222 sources and sinks

Radon-222 has been used as a tracer of sediment pore water-surface water exchange processes in a diverse range of environmental systems including rivers (McCallum et al., 2012), the coastal ocean (Burnett and Dulaiova, 2003; Dulaiova et al., 2008), and in small glacier catchments (Kies et al., 2011; Bhatia et al., 2011; Kies et al., 2015). In the following discussion, we explore the utility of  $^{222}\text{Rn}$  in tracing and quantifying meltwater fluxes from the subglacial distributed system at a large Greenland outlet glacier, Leverett Glacier. There are a number of sources and sinks capable of modulating  $^{222}\text{Rn}$  activities in a proglacial river. We use a mass balance approach, similar to those employed in studies of submarine groundwater discharge to the coastal ocean (Burnett and Dulaiova, 2003; Dulaiova et al., 2008), in order to quantify the sources and sinks for  $^{222}\text{Rn}$  in the proglacial river (Fig. 3):

$$J_{riv} = J_{dis} + J_{cha} + P_{SSL} + \lambda^{226}\text{Ra} - J_{atm} - \lambda^{222}\text{Rn} \quad (2)$$

Sources of  $^{222}\text{Rn}$  include production from  $^{226}\text{Ra}$  associated with suspended sediments ( $P_{SSL}$ ), production of  $^{222}\text{Rn}$  through the



**Fig. 4.** Average contributions from 2011 and 2012 of  $^{222}\text{Rn}$  sources to  $J_{riv}$  in the early melt season (river discharge  $<100$  m $^3$  s $^{-1}$ ) and during the late melt season (river discharge  $>100$  m $^3$  s $^{-1}$ ). Our mass-balance model suggests that throughout the melt season, the bulk of  $^{222}\text{Rn}$  in the proglacial river is derived from the distributed system. Model calculations suggested that the  $^{222}\text{Rn}$  contribution from  $P_{SSL}$ ,  $\lambda^{226}\text{Ra}$ , and  $J_{cha}$  were often  $<<1\%$  of the total  $J_{riv}$ ; in these instances, for the purpose of clarity in this figure, they have been rounded to their upper limit estimates (1%).

decay of dissolved  $^{226}\text{Ra}$  ( $\lambda^{226}\text{Ra}$  where  $\lambda$  is the decay constant for  $^{222}\text{Rn}$ ),  $^{222}\text{Rn}$  diffusion through subglacial channel sediments ( $J_{cha}$ ), and distributed system meltwater ( $J_{dis}$ ). Radon-222 sinks include radioactive decay ( $\lambda^{222}\text{Rn}$ ) and atmospheric evasion across the water/air interface ( $J_{atm}$ ). Finally,  $J_{riv}$  is the  $^{222}\text{Rn}$  flux (dpm s $^{-1}$ ) exported to the glacier's front via the proglacial river and is derived by combining our continuous  $^{222}\text{Rn}$  measurements (dpm m $^{-3}$ ) with the discharge record (m $^3$  s $^{-1}$ ). All source/sink terms can be directly evaluated except for  $J_{dis}$ ; hence we use the “flux by difference” approach (Charette et al., 2008), which assumes that the unaccounted for  $^{222}\text{Rn}$  in the mass balance model must be due to distributed system meltwater ( $J_{dis}$ ). We discuss and evaluate each source and sink term for Eq. (2) in the following sections. To provide context for the various source and sink terms below, note that  $J_{riv}$  ranged from  $3.4$ – $4.2 \times 10^6$  dpm s $^{-1}$  and  $7.2 \times 10^3$ – $3.4 \times 10^6$  dpm s $^{-1}$ , for 2011 and 2012, respectively.

#### 4.1.1. Suspended sediment $^{226}\text{Ra}$ ( $P_{SSL}$ )

Suspended sediments are a potential source of  $^{222}\text{Rn}$  to the river through decay of sediment bound  $^{226}\text{Ra}$ . From the laboratory equilibration experiments described above (Section 2.4) we determined that the surface-bound  $^{222}\text{Rn}$  activity of sediments at secular equilibrium is 0.076 dpm g $^{-1}$  (Section 3.4). To calculate  $P_{SSL}$ , we first assume that suspended sediments could produce  $^{222}\text{Rn}$  for 1–18 hours, the range of transit times observed for surface meltwater traveling through channelized drainage in the Leverett Glacier catchment (Chandler et al., 2013). Coupling this result with SSC measurements, and assuming that bedload contributed an additional 30–60% of sediment (Cowton et al., 2012), we estimated the upper and lower bounds of the contribution from  $P_{SSL}$  to  $J_{riv}$ . Over the course of each melt season,  $P_{SSL}$  supplied between  $\sim 100$  and  $2.1 \times 10^4$  dpm s $^{-1}$   $^{222}\text{Rn}$  in 2012 and  $\sim 100$  and  $2.4 \times 10^4$  dpm s $^{-1}$   $^{222}\text{Rn}$  in 2011. On average, the upper limit of our  $P_{SSL}$  estimate contributed on average  $\sim 1\%$  of  $J_{riv}$  in 2011 and 2012, respectively (Fig. 4).

#### 4.1.2. Dissolved $^{226}\text{Ra}$ ( $\lambda^{226}\text{Ra}$ )

The proglacial river also carried dissolved  $^{226}\text{Ra}$ , which could have supported  $^{222}\text{Rn}$  via its decay. We observed  $^{226}\text{Ra}$  activities of 0.02–0.09 dpm L $^{-1}$  (avg 0.04 dpm L $^{-1}$ ,  $n = 21$ ) in the proglacial river, with higher values in the early season and lower values in the late season. To solve Eq. (2) for  $J_{dis}$ , we used the average  $^{226}\text{Ra}$  activity measured in the proglacial river. Based on these results,  $^{226}\text{Ra}$  decay supplies  $\sim 400$  dpm s $^{-1}$  when river discharge was  $10$  m $^3$  s $^{-1}$  and up to  $3.2 \times 10^4$  dpm s $^{-1}$  during the maximum river discharge observed in 2012. On average during the 2011 and 2012 field seasons, dissolved  $^{226}\text{Ra}$  supplies  $\sim 1\%$  of  $J_{riv}$  (Fig. 4).

#### 4.1.3. Diffusive flux of $^{222}\text{Rn}$ in channels ( $J_{cha}$ )

The diffusive flux of  $^{222}\text{Rn}$  from channel floor sediments (subglacial or proglacial) is a potential source of  $^{222}\text{Rn}$ , particularly after the onset of widespread melting across the catchment and development of a channelized system. Our laboratory sediment diffusion experiment yielded a diffusive flux of  $0.006 \pm 0.002 \text{ dpm m}^{-2} \text{ s}^{-1}$   $^{222}\text{Rn}$  from glacial sediments. We use this result and an estimate of channel floor area during peak river discharge to quantify the potential upper limit of  $J_{cha}$ . To calculate channel floor area, we assume that all discharge moved through semi-circular channels, that channels extended to 41 km from the ice margin (Chandler et al., 2013), that the average number of channels per km catchment width was four (Schoof, 2010; Werder et al., 2013), and that channel density linearly tapered to zero between the ice sheet margin and 41 km. Furthermore, we assumed that catchment width averaged 40 km (Bartholomew et al., 2011a, 2011b) and that water moved through channels at  $3 \text{ m s}^{-1}$  (Cowton et al., 2013). Using these constraints, we calculated a channel floor area of  $2.75 \text{ km}^2$  during peak river discharge in 2012 and  $1.5 \text{ km}^2$  during peak river discharge in 2011. This estimate amounts to 0.3–0.5% of the glacier's catchment area, which is consistent with models of the channelized system (Schoof, 2010; Werder et al., 2013). Using these estimates of channel floor area,  $J_{cha}$  supplied  $\sim 6 \times 10^3$ – $1.2 \times 10^4 \text{ dpm s}^{-1}$   $^{222}\text{Rn}$  in 2011 and  $1.1 \times 10^4$ – $2.2 \times 10^4 \text{ dpm s}^{-1}$   $^{222}\text{Rn}$  in 2012. Finally, we assume that  $J_{cha}$  is only important after river discharge surpasses  $100 \text{ m}^3 \text{ s}^{-1}$  as before this time the channelized system is undeveloped (Cowton et al., 2013), and hence,  $^{222}\text{Rn}$  diffusion through channel floors is negligible. Based on these estimates,  $J_{cha}$  could account for no more than <1–10% of  $J_{riv}$  in 2011 and 2012 (Fig. 4).

#### 4.1.4. Gas exchange ( $J_{atm}$ )

The degassing of  $^{222}\text{Rn}$  out of water is a function of molecular diffusion produced by the activity gradient at the water/air interface as well as turbulent transfer, which is governed by physical processes such as wind speed, current velocity, and topography.  $J_{atm}$ , which we define as the area-normalized flux of  $^{222}\text{Rn}$  across the river/air boundary, can be written as:

$$J_{atm} = k(C_w - \alpha C_{atm}) \quad (3)$$

where  $C_w$  is the  $^{222}\text{Rn}$  activity of water,  $C_{atm}$  is the  $^{222}\text{Rn}$  activity in air (assumed here to be negligible relative to the water activity),  $\alpha$  is Ostwald's solubility constant and  $k$  is the gas transfer velocity. The gas transfer velocity is dependent on kinematic viscosity, molecular diffusion, and turbulence and is determined based on empirical relationships observed in different environments for different gases. Borges et al. (2004) suggested that the gas transfer velocity  $k$  should be in the range of 3–7  $\text{cm h}^{-1}$  while Dulaiova and Burnett (2006) calculated a gas transfer velocity for  $^{222}\text{Rn}$  up to  $10 \text{ cm h}^{-1}$  in moving water and high winds.

Because of occasional strong winds and a river current of  $\sim 1$ – $3 \text{ m s}^{-1}$  (Cowton et al., 2013), we chose a constant, upper limit  $k$  of  $12 \text{ cm h}^{-1}$  for the duration of the time series. With these assumptions,  $J_{atm}$  varied between <0.01–0.7  $\text{dpm m}^{-2} \text{ s}^{-1}$  in 2011 and 0.03–0.6  $\text{dpm m}^{-2} \text{ s}^{-1}$  in 2012. Scaling these area normalized  $J_{atm}$  values to total  $J_{atm}$  requires an estimate of river surface area ( $1100$ – $6000 \text{ m}^2$ ) between the ice terminus and the sampling site. From this surface area we determined that  $J_{atm}$  was in the range of  $800$ – $4000 \text{ dpm s}^{-1}$   $^{222}\text{Rn}$ , which was <1% of  $J_{riv}$  on average. The negligible effect of gas loss suggested by these calculations is supported by discrete  $^{222}\text{Rn}$  samples collected at the glacier's terminus, which were within error of the continuous measurements made 1 km downstream (Fig. S1; Supplementary Material).

These estimates do not account for any potential loss of  $^{222}\text{Rn}$  into the headspace of subglacial air-filled cavities. However, if these environments are largely closed systems,  $^{222}\text{Rn}$  build up in the

headspace would reduce the water–air concentration gradient and therefore minimize the  $^{222}\text{Rn}$  loss from the water phase. While there is evidence for open system channels at the ice bed within several km from the ice margin (Chandler et al., 2013), subglacial gas loss is likely much smaller than in the proglacial river. Air-filled subglacial channels far from the ice sheet margin will only exist during the falling limb of the hydrograph following substantial surface meltwater inputs and channel expansion. These cavities will close within hours to days of opening because of glacial creep (Meierbachtol et al., 2013). If large quantities of  $^{222}\text{Rn}$  were lost to air-filled cavities, then  $^{222}\text{Rn}$  activities would decrease on the falling limb of the hydrograph yet we generally observed the opposite trend in 2012 (Fig. 2). Based on our calculations and field measurements (Supplementary Material), we determined that  $J_{atm}$  was negligible in our calculation of  $J_{dis}$ .

#### 4.1.5. Radon-222 decay ( $\lambda^{222}\text{Rn}$ )

When distributed system meltwater discharges into the channelized system far from the ice margin, some fraction of  $^{222}\text{Rn}$  will decay before reaching the ice terminus. For example, during a typical 7 hour transit through the channelized system of Leverett Glacier in July (Cowton et al., 2013),  $\sim 5\%$  of unsupported  $^{222}\text{Rn}$  will decay. During the early melt season, when the snow line is at a low elevation and river discharge is  $< 10 \text{ m}^3 \text{ s}^{-1}$ , meltwater transit times in channelized drainage are likely no more than 3 h as water takes a more direct route to the margin (Chandler et al., 2013; Cowton et al., 2013). In this case, 2% of the unsupported  $^{222}\text{Rn}$  decays in the channelized system before reaching the ice margin. By the peak melt season, tracer experiments indicate that transit times in the channelized drainage system are 10–24 hours since the catchment extends upwards of  $\sim 80 \text{ km}$  from the margin and subglacial flow paths are more convoluted. As the location of these experiments was likely near the upper limit of channelized drainage in the catchment (Chandler et al., 2013), we assume nearly all meltwater traveling via channelized drainage reaches the ice sheet margin in  $< 24 \text{ h}$ . Hence for our model, we assume meltwater moving through the channelized system has a transit time between 1 and 24 hours representing a <1–17% loss of  $^{222}\text{Rn}$ .

## 4.2. Quantifying the distributed system flux

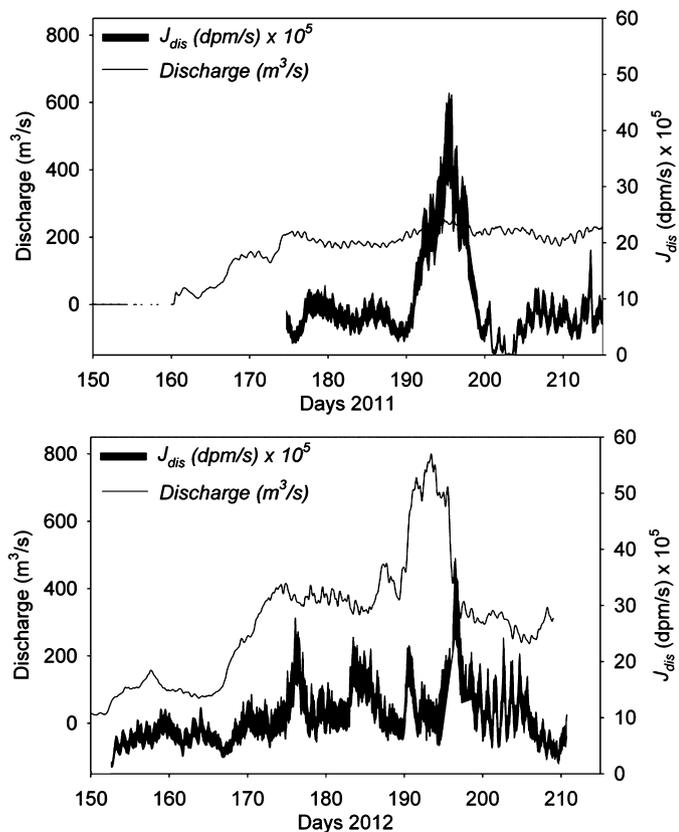
After rearranging Eq. (2), we solved for the upper and lower bounds of  $J_{dis}$  (Fig. 5).

$$J_{dis} = J_{riv} - J_{cha} - P_{SSL} - \lambda^{226}\text{Ra} + \lambda^{222}\text{Rn} \quad (4)$$

To calculate the likely range of  $J_{dis}$ , we propagated the uncertainty of each  $^{222}\text{Rn}$  source and sink. The largest source of uncertainty in  $J_{dis}$  came from the  $^{222}\text{Rn}$  and river discharge measurements used to calculate  $J_{riv}$ .

With the exception of days 200–202 in 2011 when  $J_{riv}$  dropped to near zero, non-distributed system  $^{222}\text{Rn}$  sources cannot account for the vast majority of  $J_{riv}$  (Fig. 4), we conclude that  $J_{dis}$  must contribute the bulk of the  $^{222}\text{Rn}$  flux in the proglacial river. Furthermore,  $^{222}\text{Rn}$  decay during subglacial transit through the channelized system ( $\lambda^{222}\text{Rn}$ ) and gas loss to the atmosphere ( $J_{atm}$ ), did not significantly impact the timing or flux of  $J_{riv}$ . Given that  $J_{dis}$  dominates the  $^{222}\text{Rn}$  mass balance for the Leverett Glacier proglacial river, we conclude that  $^{222}\text{Rn}$  can be used as a passive tracer of distributed system flows to the ice margin at this field site. This approach should be applicable to other settings, though an essential requirement is subglacial hydrological systems that discharge into a single proglacial river, which permits the quantification of  $J_{riv}$ , the main term in the model.

The distributed system meltwater flux ( $Q_{dis}$ ) can be calculated if  $J_{dis}$  and the  $^{222}\text{Rn}$  activity of distributed system meltwater ( $Rn_{dis}$ ) are known:



**Fig. 5.** The estimated subglacial distributed system flux ( $J_{dis}$ ) from Leverett Glacier and discharge of the proglacial river in 2011 and 2012 (black line). The width of  $J_{dis}$  represents uncertainty in this parameter (see text).

$$Q_{dis} = J_{dis}/Rn_{dis} \quad (5)$$

This approach assumes that distributed system meltwaters originate from material similar to the proglacial sediments we used to estimate  $Rn_{dis}$  (Sections 2.4 and 3.4), and that the transit time of distributed system meltwaters are  $>20$  days (Niu et al., 2015). If the transit time is shorter,  $Rn_{dis}$  would decrease and the  $Q_{dis}$  fluxes below would increase proportionately.

If the  $^{222}\text{Rn}$  activities in distributed system sediments are  $270\text{--}530 \text{ dpm L}^{-1}$  (Sections 2.4 and 3.4), then  $Q_{dis}$  would vary between  $<0.1$  to  $17 \text{ m}^3 \text{ s}^{-1}$  and  $<0.1$  to  $14 \text{ m}^3 \text{ s}^{-1}$  over the course of the 2011 and 2012 melt seasons, respectively. As a fraction of total river discharge in 2011,  $Q_{dis}$  peaked at  $\sim 22\%$  in the early season when river discharge was  $\sim 1 \text{ m}^3 \text{ s}^{-1}$  and reached a minimum of  $<0.1\%$  between days 202–204. In 2012,  $Q_{dis}$  was between 3–5% ( $0.1\text{--}0.5 \text{ m}^3 \text{ s}^{-1}$ ) of total river flow in the early season (days 130–150) and 1–4% ( $0.1\text{--}33 \text{ m}^3 \text{ s}^{-1}$ ) following day 150. The weighted mean of  $Q_{dis}$  relative to total river discharge was between 1–2.4% ( $0.35\text{--}8.5 \text{ m}^3 \text{ s}^{-1}$ ) in 2011 and 0.7–1.6% ( $<1\text{--}12.8 \text{ m}^3 \text{ s}^{-1}$ ) in 2012. However, since we were not able to obtain samples of distributed system water directly via borehole sampling (Tranter et al., 1997; Andrews et al., 2014), these estimates of  $Q_{dis}$  carry significant uncertainty; therefore, we will use  $J_{dis}$  for determining the timing and relative magnitude of distributed system fluxes.

#### 4.3. Seasonal and interannual variability in the distributed system flux

Channels at the ice sheet bed are zones of low-pressure relative to the surrounding distributed system (Röthlisberger, 1972) and generally act to scavenge distributed system meltwater (Boulton et al., 2009). However, during large surface runoff events, such as supraglacial lake drainages or periods of rapid warming, channels

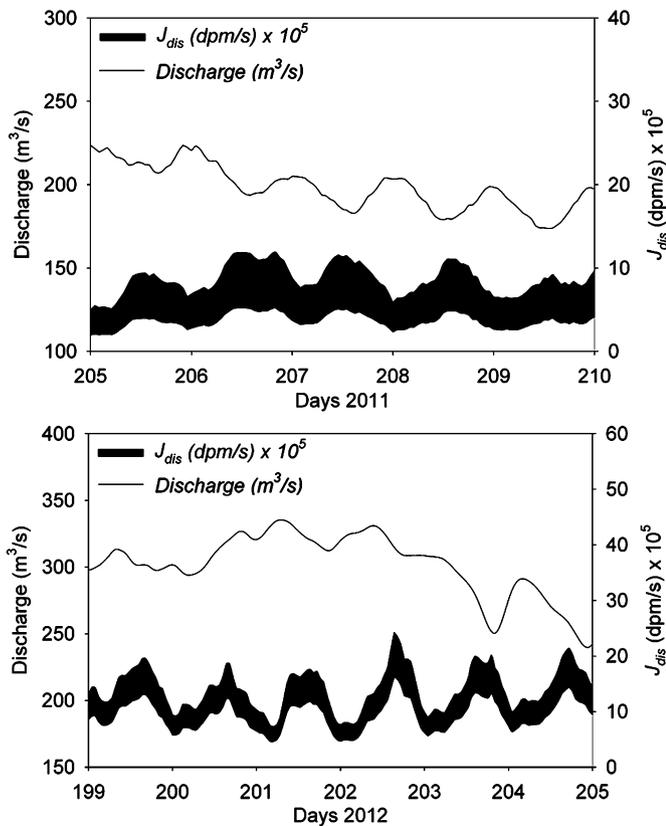
may be overwhelmed forcing water into the distributed system (Bartholomew et al., 2012; Gulley et al., 2009). Once surface meltwater runoff decreases, water pressure in channels falls and the flux of distributed system drainage to the ice sheet margin surges (Hubbard et al., 1995; Boulton et al., 2009). This process has been hypothesized to result in more connectivity between channelized and distributed systems causing an overall increase in the spatial extent of subglacial drainage (Andrews et al., 2014). This in turn may lead to less water volume at the ice sheet bed and could explain observed mid-late summer slowing of land-terminating sections of the GrIS (Sole et al., 2013). Hence, the mechanisms that control the characteristics of distributed system drainage likely play a key role in modulating the speed of GrIS outlet glaciers, especially because distributed system networks make up by far the largest portion of the ice sheet bed area.

In 2011, the largest multiday peak in  $J_{dis}$  (Fig. 5) (days 190–200) occurred during the onset of melting and ice acceleration (Sole et al., 2013) at high elevations ( $>1400 \text{ m}$ ) within the catchment, and the expansion of the channelized system to at least 40 km from the ice sheet margin (Chandler et al., 2013). During the 2012 time series, there were four major multiday peaks in  $J_{dis}$  (Fig. 5). The largest 2012 peak occurred on the falling limb of the hydrograph of the widely publicized extreme melting event (Nghiem et al., 2012) during which river discharge reached  $\sim 800 \text{ m}^3 \text{ s}^{-1}$  (day 196; Fig. 5). These results provide direct evidence that drainage of distributed regions follows periods of channelized system expansion due to rapid increases in surface meltwater runoff. Following the largest peaks in  $J_{dis}$  in 2011 and 2012, significant diurnal variations were observed in  $J_{dis}$  (days 205–210 in 2011 and days 199–205 in 2012; Fig. 6). These daily cycles in  $J_{dis}$  suggest a high degree of connectivity between the distributed and channelized systems with increases in flux from the distributed on the falling limb of the hydrograph when subglacial water pressure subsides (Fig. 6).

Bartholomew et al. (2011a, 2011b) and Butler (2014) found evidence that SSC/EC peaks at Leverett Glacier are triggered by supraglacial lake drainage events. These events likely increase the connectivity of the channelized system (Bartholomew et al., 2011a), and may lead to the expulsion of distributed system meltwater. In general, peaks in  $J_{dis}$  were not correlated with SSC/EC peaks, nor did peaks in SSC/EC lead to enhanced  $^{222}\text{Rn}$  concentrations (Fig. 2). Lake drainage events clearly increase the suspended sediment load which may lead to post mixing solute acquisition reactions, causing meltwaters to rapidly acquire dissolved solutes (Raiswell, 1984; Tranter, 1993). Conversely,  $^{222}\text{Rn}$  equilibrium in the distributed system will likely be reached long before subglacial meltwaters become saturated with respect to weathering minerals. Hence, we would expect more variation in solute concentrations (EC) in the distributed system than  $^{222}\text{Rn}$  activities. Consequently, lake drainage events could produce the observed SSC/EC peaks without a corresponding  $J_{dis}$  peak if they act to flush small volumes of distributed system meltwater with high solute concentrations to the ice sheet margin.

## 5. Conclusions

Using a mass balance model for  $^{222}\text{Rn}$  in a large glacial catchment of the GrIS, we found that on average,  $>90\%$  of the  $^{222}\text{Rn}$  in the proglacial river is sourced from the subglacial distributed system. Hence, at Leverett Glacier,  $^{222}\text{Rn}$  acts as a conservative, passive tracer of distributed system meltwater fluxes. These fluxes varied on diurnal, seasonal, and interannual time scales. Based on  $^{222}\text{Rn}$  measurements, large peaks in distributed system drainage appear to be initiated by the expansion of the channelized system into presumably distributed regions of the ice sheet bed and by rapid increases in supraglacial meltwater runoff. During a large



**Fig. 6.** Diurnal variations in  $J_{dis}$  occurred in both the 2011 and 2012 melt seasons. In general,  $J_{dis}$  peaks occurred on the falling limb of the hydrograph. We hypothesize that during this period, distributed system meltwater was drawn into the channelized system as water pressure dropped in subglacial channels.

multiday  $J_{dis}$  peak in 2011 (days 190–200; Fig. 5), SF<sub>6</sub> tracer experiments (Chandler et al., 2013) and ice acceleration (Sole et al., 2013) suggested the channelized network expanded coincident with the  $J_{dis}$  peak. In 2012, four major multiday  $J_{dis}$  peaks were observed (Fig. 5), the biggest of which occurred on the falling limb of the hydrograph during the largest surface meltwater runoff event observed in Greenland since at least 1889 (days 196; Nghiem et al., 2012). These results imply that rapid warming events, which initially cause short term glacial acceleration (Tedstone et al., 2013), may lead to enhanced distributed system drainage, a process which could lessen the total water volume at the ice sheet bed and ultimately, explain the observed mid-summer ice sheet slowing at Leverett Glacier (Sole et al., 2013; Tedstone et al., 2013). Following the largest peaks in  $J_{dis}$  in 2011 and 2012, significant diurnal variations were observed in  $J_{dis}$ , indicative of a highly connected distributed system whereby the distributed system water flux substantially increases at night when channelized system water pressure subsides.

Based on our laboratory-based sediment equilibration measurements of <sup>222</sup>Rn activities in distributed system fluids, we estimate that distributed system meltwater fluxes vary seasonally and are on the order of 1–5% of river discharge. The weighted mean  $Q_{dis}$  relative to total river discharge was between 1–2.4% in 2011 and 0.7–1.6% in 2012. Future studies should endeavor to collect samples for <sup>222</sup>Rn analysis directly from distributed and channelized systems so as to better constrain  $Q_{dis}$ . Furthermore, utilizing continuous <sup>222</sup>Rn measurements provides a practical tool to capture hourly variations in <sup>222</sup>Rn activity as well as episodic events that might otherwise be missed if solely relying on discrete <sup>222</sup>Rn measurements. Additionally, the detection limit and measurement uncertainty using the water probe extraction method (Section 2.3) is much lower than discrete sampling (Supplementary Material,

Fig. S1). Our results demonstrate that there is great potential for continuous <sup>222</sup>Rn measurements in proglacial rivers to aid our understanding of how distributed system fluxes impact glacial hydrology, ice-dynamics, and biogeochemical fluxes.

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### Appendix A. Supplementary material

Supplementary material related to this article can be found online at <http://dx.doi.org/10.1016/j.epsl.2016.12.039>.

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