

559 **Supplementary Material**

560 *S1 Water probe response time*

561 During the 2011 and 2012 field seasons, several factors related to the water probe
562 setup may have influenced the response time of the water probe. First, in order to keep
563 the RAD7 a safe distance from the unstable riverbank, the length of tubing between the
564 detector and water probe had to be increased (from 8 to 12.5 m) effectively changing the
565 system air volume from 1650 to 1750 cm³. Furthermore, the interface area of the
566 membrane coil changed as two Accruel® membrane coils of slightly different lengths (a
567 2.1 m coil and the 2.2 m coil within the Durrige Inc. ‘water probe’) were used
568 interchangeably across the two field seasons. The effect of these changes was quantified
569 using the empirical relationships developed by Shubert et al., (2012), which suggest that
570 our minor changes in air volume and membrane interface area likely only caused a 12-
571 minute difference in equilibration time. This is a negligible difference when considering
572 that the minimum time-scale over which significant changes in observed river ²²²Rn were
573 diurnal. Furthermore, a field test was conducted in which both membrane coils were
574 simultaneously deployed in the proglacial river for 12 hours. During this test, both
575 systems recorded ²²²Rn activities well within the statistical counting errors. Consequently,
576 the continuous ²²²Rn record reported here is a compilation of results obtained using both
577 water probes. During deployment, the membrane coil was checked daily for wear and
578 sediment buildup on its surface. Throughout either field seasons, no algae or sediment
579 buildup was observed. To test the effectiveness of long term deployment of the
580 membrane coil over the course of a melt season, two probes were deployed
581 simultaneously, one that had been in continual use for ~30 days and a second unit with a

582 new membrane coil. Following an initial equilibration period, both units measured
583 identical ^{222}Rn activities (within the method's uncertainty) for several days.

584 To assess the water probe's response to changes in ^{222}Rn activity, we conducted
585 several laboratory experiments. First, the water probe was deployed simultaneously with
586 the more conventional air-water equilibrating spray chamber (Burnett and Dulaiova,
587 2003) in a 200 L tank of circulating 10°C seawater for 70 hours. Seawater was
588 continuously pumped into the tank from Vineyard Sound, ~50 m offshore of Woods Hole
589 (MA). The air-water equilibrator spray chamber provided a baseline from which to
590 compare the water probe, because with an optimum setup, its equilibration time is less
591 than 30 minutes (Schubert et al., 2012). The residence time of seawater in the tank was
592 <1 hour and water was kept in constant motion using six submersible bilge pumps, each
593 capable of pumping $\sim 30 \text{ L min}^{-1}$. Slow but significant changes in ^{222}Rn were observed by
594 both the spray chamber and water probe likely caused by the changing tide and submarine
595 groundwater discharge (Burnett and Dulaiova, 2003). The equilibration time was defined
596 as the length of time at which the recorded ^{222}Rn activities reached an activity plateau
597 within the statistical counting errors. With this experimental setup, the water probe
598 required an initial six-hour equilibration time while the spray chamber reached
599 equilibrium in <30 min. Subsequent changes in ^{222}Rn activity measured by the water
600 probe lagged 1-2 hours behind the spray chamber. In a separate experiment, the water
601 probe and spray chamber were allowed to equilibrate with ^{222}Rn -free water before being
602 moved quickly into a tank containing ^{222}Rn -enriched groundwater. In this case, both
603 water probe and spray chamber systems responded to the activity change in <30 minutes
604 though the water probe required much longer to reach the new equilibrium plateau.

605 To determine the equilibration time of the water probe system in the proglacial
606 river, we examined the first six hours of data recorded after the water probe was freshly
607 deployed (see Figure S2 for one example). Our analysis included 14 separate
608 deployments in 2011 and 2012 in river flows ranging from 1 to 750 m³ s⁻¹. In each case,
609 an equilibration plateau was reached within two hours of deployment regardless of river
610 flow rate or system configuration (as described above). The equilibration time was
611 therefore three times faster than the laboratory experiments. This was likely because
612 water in the proglacial river was flowing much faster than in laboratory experiments
613 keeping the ²²²Rn activity gradient at the water/air interface of the membrane coil closer
614 to 100%. These results support the findings of Hofmann et al. (2011) and Schubert et al.
615 (2012) showing that the water flow rate over the membrane coil is the most important
616 factor for passive ²²²Rn extraction. Because of the much faster equilibration time in the
617 proglacial river, we expect the water probe's response time to changing ²²²Rn activities
618 was also faster in the field than the one to two hours suggested by laboratory experiments.

619 In summary, when interpreting results from continuous ²²²Rn measurements, we
620 assume changes in ²²²Rn activity recorded by our methods occurred within one hour of
621 actual ²²²Rn activity changes in the proglacial river. Also, we have excluded ²²²Rn results
622 from the first two hours from each fresh deployment while the water probe was
623 equilibrating.

624 S2 Sediment Properties

625 The porosity and bulk density of glacial flour collected in the proglacial river was
626 determined using the moisture content and particle density measured in the laboratory.

627 Moisture content (%M) was determined by weighing sediments before and after drying at
628 100°C: using Equation S1:

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$$\%M = \frac{\text{wet wt.} - \text{dry wt.}}{\text{wet wt.}} \times 100$$

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(S1)

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Grain density (ρ_s) was determined using the oven dry weight and volume of sediment.

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The volume of sediment was determined by adding the sediments to a volumetric flask

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and measuring the weight of water displaced by the sediments.

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Bulk density (β_D) was calculated using Equation S2

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$$\beta_D = \frac{1}{\left(\left[\frac{1}{100 - \%M}\right] \times 100 + \frac{1}{\rho_z}\right) - 1} \quad (S2)$$

637

where ρ_s is the average sediment grain density of triplicate analysis. Finally, porosity (ϕ)

638

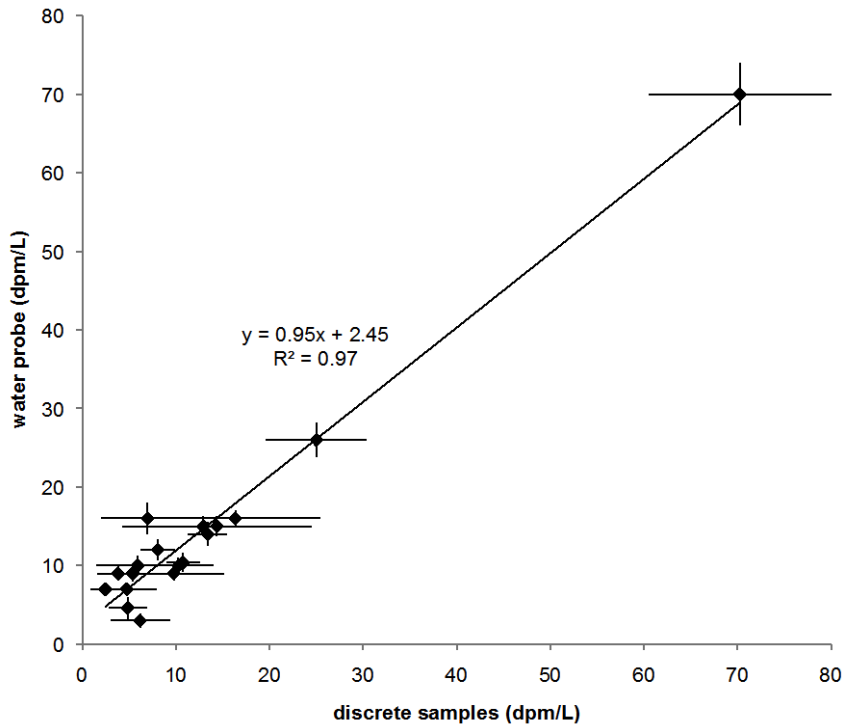
was estimated from Equation S3.

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$$\phi = (\rho_s - \beta_D) / \rho_s \quad (S3).$$

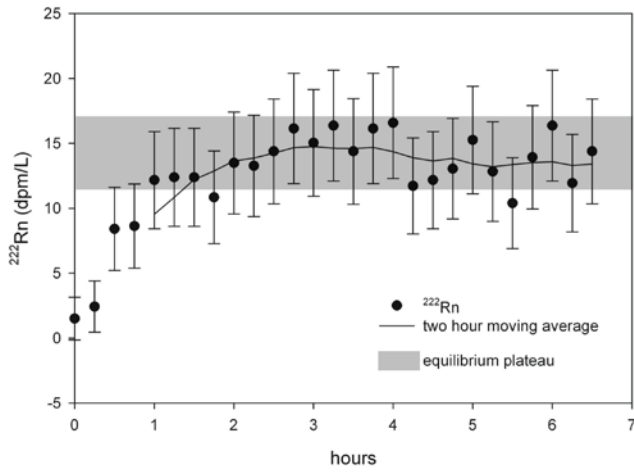
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643 Figure S1: Comparison of discrete ^{222}Rn samples with comparable time-series
 644 measurements using the water probe. Error bars represent 1-sigma counting errors on
 645 each measurement. Many of these discrete ^{222}Rn samples were taken at the ice terminus
 646 while the water probe measured ^{222}Rn downstream. This implies that gas loss in the
 647 proglacial river between the ice terminus and the water probe was within the errors of our
 648 measurements.

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651 Figure S2: Equilibration time of the ^{222}Rn water probe on 6/2/2011 (day 153) after being
 652 deployed in the proglacial river. Throughout the 2011 and 2012 field seasons, the water
 653 probed required 1-2 hours to reach an equilibrium plateau, the time at which the recorded
 654 radon activities reach an equilibrium activity plateau within the statistical counting errors.

655

656 **Table S1.** Summary of 250 mL discrete ^{222}Rn samples collected in the proglacial river in
 657 2011.

Distance	Day	Date	^{222}Rn	+/-	EC
			dpm L ⁻¹		$\mu\text{S cm}^{-1}$
0.5	128.5	5/8/11	15.9	6.4	
0.5	129.6	5/9/11	3.2	2.1	
0.5	132.5	5/12/11	3.2	2.8	69
0.5	132.5	5/12/11	7.0	5.4	69
0.5	132.8	5/12/11	4.3	3.0	75
0.5	132.8	5/12/11	5.9	4.4	75
0.5	133.4	5/13/11	4.8	2.1	94
0.5	133.4	5/13/11	7.0	6.4	94
0.5	133.7	5/13/11	18.2	10.1	79
0.5	133.7	5/13/11	14.5	7.5	89
0	133.6	5/13/11	4.3	3.9	
0	133.6	5/13/11	13.4	2.1	
0.5	134.5	5/14/11	9.1	2.1	90
0.5	134.5	5/14/11	7.0	4.4	90
0.5	135.5	5/15/11	5.4	2.8	74
0.5	135.5	5/15/11	7.0	6.2	76
0.5	136.0	5/16/11	10.8	1.8	81
0.5	137.5	5/17/11	9.1	6.2	76
0.5	137.5	5/17/11	1.6	2.0	76
0.5	139.6	5/19/11	4.3	1.8	81
0.5	140.8	5/20/11	5.9	1.1	73
0.5	140.8	5/20/11	5.4	2.8	73
0.5	141.6	5/21/11	14.0	4.1	75
0.5	145.8	5/25/11	4.3	4.7	65
0.5	144.6	5/24/11	6.9	3.2	88
0	147.7	5/27/11	67.4	11.6	49
0	147.7	5/27/11	32.2	7.3	45
0.5	147.8	5/27/11	7.5	2.8	45
0.5	147.8	5/27/11	12.9	3.0	45
0	148.5	5/28/11	41.4	8.1	85
0	148.5	5/28/11	54.3	7.3	84
0.5	148.5	5/28/11	70.3	9.7	62
0.5	149.8	5/29/11	24.3	6.2	40
0	149.8	5/29/11	17.8	8.8	40
0	151.9	5/31/11	17.8	3.1	37
0	148.8	5/28/11	6.5	3.5	61
0	150.8	5/30/11	5.4	1.3	40

658 0.5 150.8 5/30/11 14.7 6.8 40

Distance	Day	Date	²²² Rn	+/-	EC
			dpm L ⁻¹		μS cm ⁻¹
0.5	153.7	6/2/11	22.6	9.5	36
0	153.7	6/2/11	3.2	2.8	36
0	152.7	6/1/11	24.1	7.0	36
0.5	156.6	6/5/11	5.4	3.7	14
0.5	156.6	6/5/11	4.1	4.8	15
0.5	156.7	6/5/11	17.1	3.0	12
0.5	156.7	6/5/11	17.2	5.3	12
0	157.7	6/6/11	4.3	3.0	44
0	157.7	6/6/11	6.5	5.3	45
0.5	158.3	6/7/11	4.8	3.1	21
0.5	159.2	6/8/11	3.8	2.1	37
0.5	159.3	6/8/11	9.7	5.4	36
0.5	153.7	6/2/11	8.2	5.7	42
0.5	153.7	6/2/11	20.5	5.5	50
0.5	162.7	6/11/11	1.6	1.1	16
0.5	162.7	6/11/11	3.3	1.2	16
0.5	165.0	6/14/11	4.9	6.9	35
0.5	165.5	6/14/11	5.4	2.1	27
0	166.7	6/15/11	3.2	2.1	23
0	166.7	6/15/11	10.7	5.8	23
0.5	167.5	6/16/11	2.7	1.1	18
1	167.6	6/16/11	2.7	3.2	18
1	170.0	6/19/11	3.8	1.1	12
1	170.0	6/19/11	4.3	1.8	12
0	172.7	6/21/11	2.7	2.7	14
0	172.7	6/21/11	4.8	4.7	14
0	184.6	7/3/11	2.1	0.9	
1	198.9	7/17/11	1.6	2.0	