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Seasonal controls on meteoric ^{7}Be in coarse-grained river channels

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Abstract:

Cosmogenic ^{7}Be is a natural tracer of short-term hydrological processes, but its distribution in upland fluvial environments over different temporal and spatial scales has not been well described. We measured ^{7}Be in 450 sediment samples collected from perennial channels draining the middle of the Connecticut River Basin, an environment that is predominantly well-sorted sand. By sampling tributaries that have natural and managed fluctuations in discharge, we find that the ^{7}Be activity in thalweg sediments is not necessarily limited by the supply of new or fine-grained sediment, but is controlled seasonally by atmospheric flux variations and the magnitude and frequency of bed mobilizing events. In late winter, ^{7}Be concentrations in transitional bedload are lowest, typically 1 to 3 Bq kg^{-1} as ^{7}Be is lost from watersheds via radioactive decay in the snowpack. In mid-summer, however, ^{7}Be concentrations are at least twice as high because of increased convective storm activity which delivers high ^{7}Be fluxes directly to the fluvial system. A mixed layer of sediment at least 8 cm thick is maintained for months in channels during persistent low rainfall and flow conditions, indicating that stationary sediments can be recharged with ^{7}Be. However, bed mobilizing rain on snowmelt events in late Spring can 'reset' ^{7}Be amounts and concentrations in the channel as previously buried 'old' sediment with low ^{7}Be is mixed into the thalweg. We conclude that given proper temporal and spatial sampling, ^{7}Be is a valuable tracer of seasonal-timescale mass transport and exchange in coarse-grained fluvial systems. Copyright © 2013 John Wiley & Sons, Ltd.

KEY WORDS fallout; radionuclides; sediment; beryllium; bar; channel; transport; age

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INTRODUCTION

'Fallout' radionuclides that adsorb strongly to solids have been used for decades to quantify sediment dynamics in lakes, estuaries, and coastal environments (Krishnaswami *et al*., 1980; Olsen *et al*., 1993; Sommerfield *et al*., 1999; Willenbring and von Blanckenburg, 2010). Two naturally occurring fallout radionuclides useful as sediment tracers in fluvial environments, cosmogenic ^{7}Be ($T_{1/2}$ = 53 d) and radon-derived ^{210}Pb ($T_{1/2}$ = 22 y), are deposited on the earth's surface nearly continuously as precipitation falls on soils, waters, and exposed regolith. Both Be and Pb have a strong affinity for inorganic and organic sediments (Hawley *et al*., 1986; You *et al*., 1989; Chen *et al*., 2006; Taylor *et al*., 2012), and their similar meteoric source term and contrasting half-lives make them useful over very pertinent spatial and temporal scales (Bonniwell *et al*., 1999; Aalto *et al*., 2003; Walling *et al*., 2009). ^{210}Pb, along with weapons-derived ^{137}Cs, has been extensively developed as a valuable dating tool for measuring sedimentation and transport rates over decadal timescales in rivers, lakes, floodplains, and near-shore environments (Appleby and Oldfield, 1992; Fuller *et al*., 1999; Aalto *et al*., 2003). In estuarine and coastal shelf environments, ^{7}Be is an established tracer of sediment deposition and reworking at the event or seasonal timescale (Olsen *et al*., 1986; Canuel *et al*., 1990; Sommerfield *et al*., 1999; Palinkas *et al*., 2005), but, it has been used less frequently in upland watershed studies than ^{210}Pb or weapons-derived ^{137}Cs. Gamma spectrometry allows for the convenient measurement of ^{210}Pb (and its grandparent ^{226}Ra), ^{137}Cs, and ^{7}Be simultaneously in sediment samples without chemical separation steps. Larsen and Cutshall (1981) were the first to suggest the direct quantification of ^{7}Be (without radiochemical separation) in sediments by high-resolution gamma spectrometry as a way to trace rapid sedimentation in tidal rivers and near-shore environments. They analyzed ~250 g sediment samples on co-axial type lithium-drifted Germanium (Ge) detectors that had a counting efficiency of ~1.5% at the ^{7}Be spectral region of interest (477–478 keV). Improved pure 'intrinsic'

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Ge detectors are now available with ultra low-background configurations and crystal geometries producing sensitivities considerably higher than this for a flat sample (3–8%), which has led to an increase in the number of laboratories making routine ^{7}Be measurements in recent years (e.g. Walling, 2012).

The first detailed watershed-scale study of ^{7}Be found that in an alpine basin, export was dominated by transport on silt and clay, and that the nuclide had a relatively short erosional residence time of < 1 year (Dominik *et al*., 1987). Afterwards, Bonniwell *et al*. (1999) used measurements of ^{7}Be/^{210}Pb on suspended sediments to show that fine particles are transported relatively long distances – in steps of tens of km during a snowmelt flood in the Gold Fork Watershed of Idaho, USA. In this case, ^{210}Pb and ^{137}Cs were used to normalize for the effects that particle size and source area might have on ^{7}Be concentrations, so that the ratio reflected mostly time as ^{7}Be decayed during transit downstream. Fine sediment deposition and re-suspension rates in rivers can be calculated by projecting ^{7}Be inventories that should be present in bed load based on atmospheric fallout (Jweda *et al*., 2008). This technique was used in the Fox River, in Wisconsin, to show that a polychlorinated biphenyl-contaminated sediment layer was largely re-suspended during high flow despite the impounded nature of the river (Fitzgerald *et al*., 2001).

^{7}Be can be used to measure sediment travel distances and to establish timescales of transitional bedload sequestration. Salant *et al*. (2007) repeatedly measured ^{7}Be in point bar and channel sands in a regulated river immediately downstream of a dam in Vermont, U.S.A. During winter months, sediment stored behind the dam became depleted in ^{7}Be; when the gates were opened in early spring the pulse of depleted 'new' sediment was discernible from the surrounding 'active' sediments that were supplied by tributaries and previously exposed bars. Transport rates of 30 to 80 m d^{-1} were calculated using the known starting point of the sands and monthly sampling at fixed points to monitor the sediment pulse as it moved down river. In a study of a coastal plain river in Maine, U.S.A., Fisher *et al*. (2010) used a constant initial ^{7}Be dating model to measure the timescale of sediment storage behind large woody debris and boulders where the sediment source was simple to characterize. In independently-assessed 'transport-limited' reaches of the Ducktrap River, sediment sequestration timescales behind in-stream obstructions were >100 days, but in supply-limited reaches, sediment was exchanged more quickly.

^{7}Be decays over a timescale that makes it appropriate for quantifying and tracing processes and deposits associated with a single large hydrologic event, and when combined with measurements of ^{210}Pb and ^{137}Cs, a more widely used tool for identifying different timescales of sediment transport and deposition may emerge (Walling *et al*., 2009; Walling, 2012). The use of ^{7}Be as a tracer in fluvial environments requires an understanding of how its activity changes over short temporal and spatial scales, so that ultimately, measurements of ^{7}Be can be used to evaluate sediment source, mixing, transport, and 'age' (Wallbrink *et al*., 1999; Matisoff *et al*., 2005). Until recently, ^{7}Be has largely been used as a tracer of fine-grained material, and the framework for applying radionuclides as tracers of sediment dynamics in shallow waters is largely based on the study of estuarine and coastal systems (Olsen *et al*., 1986; Dibb and Rice, 1989; Sommerfield *et al*., 1999) or on suspended sediment (Bonniwell *et al*., 1999), where 'new' and 'old' sediment are easily defined. It is less clear, however, how atmospheric flux variability and sediment transport processes might regulate ^{7}Be in more transitional environments, where sediment is being exchanged on short timescales, and whether or not methods and assumptions developed for fine-grained sediments are valid in coarse-grained systems. Here, we measure the spatial and temporal distribution of ^{7}Be in rivers draining the post-glacial landscape of New England, which are sand and gravel dominated systems with little to no clay (Rainwater, 1962; Salant *et al*., 2006). Our goal here is to provide deeper insight into how ^{7}Be can be developed as a sediment tracer in coarse-grained fluvial environments.

METHODS

Exposure studies

It is well established that soils and fine-grained

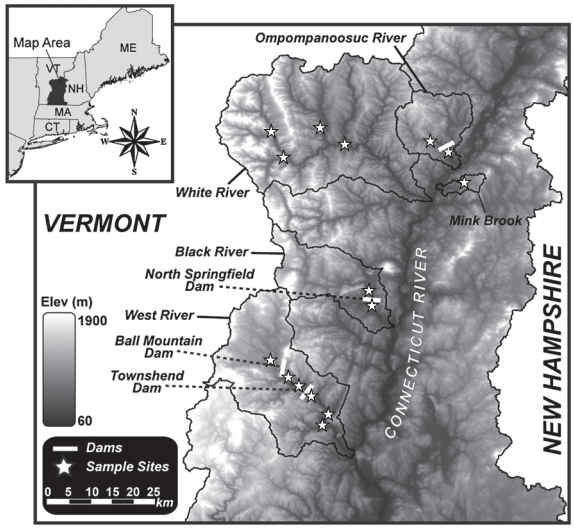

Figure 1. Watersheds (outlined in black) in New Hampshire and Vermont that were sampled for thalweg sediments on a seasonal to monthly basis from 2004 to 2008. At sampling sites below dams, sediment was collected from multiple positions downstream at a spacing of 1 to 2 km. At tributary junction sites, sediment was collected from the tributary and the mainstem above and below the junction. Further details about the sampling locations are given in the Methods

sediments exposed to atmospheric deposition will develop ^{7}Be profiles that decline approximately exponentially with depth. Measurable penetration into such surfaces is usually limited to a few cm (Wallbrink and Murray, 1996; Blake *et al.*, 1999). To test whether or not the coarser grained sediments in our environment would quantitatively retain ^{7}Be from rainfall, we collected high-resolution profiles (2-cm intervals) of sediment packages that would be delivered to the thalweg during storms. One sediment profile was collected from a point bar on the West River immediately below Ball Mountain Dam (Figure 1). Another sediment profile was collected from the middle of a hemlock-vegetated scroll bar 25 km downstream of the dam. The elevation of this sand deposit was between the one and two-year flood levels, and it had been stable for at least six months prior to sampling.

In addition to sampling quasi-stable sediment deposits *in situ*, we exposed diorite gravel to precipitation and analyzed the resulting ^{7}Be distribution in a more controlled setting. The goal here was to evaluate if fresh, relatively unweathered coarse-grained sediments would retain ^{7}Be. Using a shatterbox to break relatively unweathered diorite into small fragments, we isolated the 2–4 mm fraction using sieves. After washing the 2–4 mm sediment with de-ionized water to remove fines and rock dust, we packed it in a plastic bin (45 cm long by 20 cm wide) that had holes in the bottom for drainage. We filled the bin with the gravel to a height of 8 cm and placed it on the roof of a four-story building in Hanover, NH for 2 months during the late Spring of 2007. Weekly rainfall was collected during this period using plastic containers that were open to the atmosphere. At the end of the exposure period, we used orange spray paint to label the uppermost layer of exposed grains. This layer was then removed using tweezers, and then another coat of spray paint was applied to identify the next layer, which was then removed in the same manner. We did this multiple times so that we sampled at the grain scale (2–4 mm) from the surface downwards, and each layer was counted separately for ^{7}Be. Rainfall samples were acidified before each collection to prevent ^{7}Be adsorption to the plastic (Baskaran, 1995).

Seasonal sampling of channel sediments in larger rivers of the Connecticut River Basin

We collected sediments from the thalwegs of several regulated and unregulated tributaries of the Middle Connecticut River Basin in New Hampshire and Vermont on a monthly to seasonal basis from 2004 to2008 (Figure 1). The region receives ~ 110 cm/year of precipitation, of which ~30% is snow. Because of the crystalline bedrock and high energy environment of the last glacial retreat, soils and streams in our study watersheds have little fine-grained material. Suspended sediment concentrations are low ($<200\,mg\,l^{-1}$), and fluvial transport is generally dominated by bedload saltation of medium to coarse sand (Rainwater, 1962; Dade *et al.*, 2011). Most of the major tributaries of the Connecticut River were dammed by the early 1960s for flood control, and we use these impoundments here as an experimental dimension to study how changes in sediment supply and water discharge would affect ^{7}Be dynamics. Since the construction of these flood control structures, peak flows have been reduced with most gauged sites having a ~60% reduction in 2-year bankfull discharge (Q_2), essentially disconnecting the floodplain from the post-dam hydrologic regime and limiting channel-floodplain exchanges (Magilligan and Nislow, 2001; Nislow *et al.*, 2002). The White River is the largest unregulated watershed in the Connecticut River Basin, which we sample as a control to compare with the regulated reaches (Figure 1).

Study sites on regulated rivers were located upstream and downstream of dams on the Black, West, and Ompompanoosuc Rivers in eastern Vermont (Figure 1). The dams have different management styles, giving us a range of sediment transport conditions to study. Because of its large storage reservoir that is maintained year-round, the Black River below the North Springfield Dam (NSD) has been scouring, with the post-dam mean bed elevation dropping by about 29 cm since impoundment (Magilligan *et al.*, 2008). In contrast, the Union Village Dam (UVD) on the Ompompanoosuc River operates a storage reservoir only during the winter and early spring, which causes aggradation downstream (Salant *et al.*, 2007) as sediment passes through when the gates are opened after that period. Near each dam, we typically sampled one site upstream of the backwater effect and had at least four sampling positions downstream of the structure at ~1 to 2 km spacing. At tributary junction sites in the West and White Basins (Figure 1), we collected sediment from the main stem above the junction, below the junction, and from the smaller tributary. The regulated and unregulated sites have similar drainage areas and geology, with bedrock and till dominated by quartzite, gneiss, and schist. At all sampling positions, we collected ~0.5 kg of sediment from the thalweg over an area of approximately 20 cm $\times$ 20 cm using hand or trowel, avoiding recently deposited material from adjacent floodplains or hillsides. This material was oven dried, and the sieved ($<$ 2 mm) fraction was used for radionuclide analysis.

Monthly point bar and riffle sediment sampling in Mink Brook, NH

In addition to sampling the larger White, West, Ompompanoosuc, and Black Rivers on a seasonal to monthly basis, in 2006, we repeatedly sampled exposed point bar and submerged riffle sediments in Mink Brook, a small unregulated tributary (46.7 km^2) draining the western flanks of Moose Mountain (Figure 1). Our site,

located near a former USGS stream gage (# 01141800), has a contributing watershed size of 11.9 km^2 and an estimated Q_2 of 5.4 m^3/s. The bed is relatively steep (ranging from 1.5–2% across the reach) and is mantled with sand, gravels, and small cobbles (mean particle size is 90 mm), which is supplied as till deposits erode from adjacent steep hillsides. Here, we took monthly sediment samples in low gradient riffles along a stable reach that appeared to be neither eroding nor aggrading. After placing part of a 55-gallon steel drum in the stream to block flow, the surface armor was removed, and we sampled four discrete horizontal layers two cm thick to a total depth of 8 cm. This sampling method allowed us to retain and collect the silt and sand in each layer because flow was stopped inside the barrel. Twelve vertical profiles were collected (0–8 cm) at least one per month from March to December of 2006, with a duplicate profile collected in December to assess small-scale spatial variability. In addition to the riffle sampling here, beginning in April 2006, surface samples to 2 cm depth were collected from a nearby point bar which we considered to be a potential sediment source to the thalweg during storms. We analyzed ^{7}Be on the <2 mm sediment, and a complete grain size analysis of this fraction was done on each sample after nuclide analysis by first soaking the sample in 10% sodium hexametaphosphate for at least 2 h, then wet sieving the sample through a 62.5 micron sieve. The >62.5 micron portion was then dried completely at ~60 °C and passed through a series of nested sieves (0.125 to 1 mm) on a motorized sieve shaker, after which the sample retained on each sieve was weighed. We also measured specific sediment surface area on the Mink Brook sediments with a Micromeritics FlowSorb III 2305 surface area analyzer using a 30% N_2/70% He gas mixture. Accuracy of the instrument is ±3% with reproducibility of ±0.5%.

Analytical methods

We measured ^{7}Be activity in samples by low-background counting its 477.6 keV gamma emission. For this, we use shielded 'Broad Energy' intrinsic Germanium Detectors (BE5030 and BE3830, Canberra Industries, Meriden, CT) designed with ultra-low-background cryostats and remote detector chambers. Sample geometry for sediment samples was a 105 ml plastic dish (8.8 cm diameter, 1.7 cm thickness), and the rainfall samples were analyzed using a 700 ml Marinelli-style container. Detector efficiency for the ^{7}Be region of interest at 477–478 keV for each container was determined by counting a solution containing certified values (Eckert & Ziegler Isotope Products, Valencia, CA) of over a dozen different nuclides decaying at energies ranging from 46 keV to 1836 keV. Our counting efficiency at 477–478 keV for the 105 ml sediment container was determined to be 3.4 to 4.7%, depending on the size of the specific detector, and resolution here is approximately

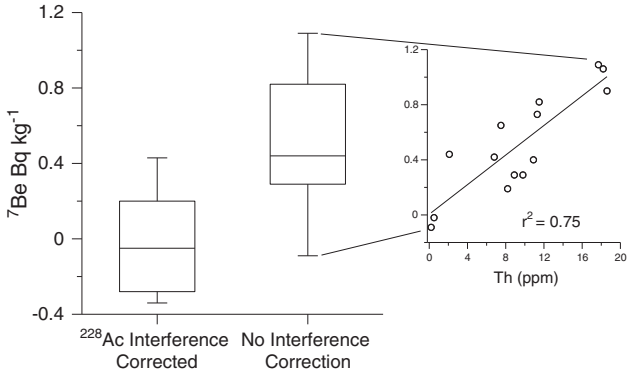

Figure 2. Box and whisker plots showing measurements of 16 different sediment samples with effectively zero ^{7}Be ('blanks') with and without correcting for the 228Ac emission at 478 keV. Inset shows correlation between the bias of uncorrected ^{7}Be measurements and Th content (Bq ^{7}Be kg^{-1} = 0.054 * ppm Th + 0.005)

1 keV peak width at half of the maximum height. Larsen and Cutshall (1981) found that for a 250 g sediment sample, differences in attenuation of the ^{7}Be gamma between a calibrating solution and sediment matrix were negligible, so we make no attenuation correction for our liquid-derived detection efficiency. We summed the counts in the five-channel ^{7}Be region of interest spanning 476.5–478.5 keV and used a best line-of-fit model background based on neighbouring channels which avoided the documented 480 keV decay emission of ^{214}Pb (Morel *et al.*, 2004; Landis *et al.*, 2012). Masses of 125–175 g (<2 mm) were typically counted for 24–48 h. Other U-series radionuclide (^{210}Pb, ^{226}Ra) and Th-series (^{228}Ac) activities were determined using standard techniques (Cutshall *et al.*, 1983; Murray *et al.*, 1987).

We carefully evaluated the detection limits for ^{7}Be in sediment by measuring 16 different samples that could be considered 'blanks'; that is, they were a range of sediment and regolith samples that had been collected from deep (>20 cm) in profiles and stored for at least 9 months. Analysis of these blanks by simply using the net photopeak area at 477–478 keV revealed a positive bias of 0.51 Bq ^{7}Be kg^{-1} (± 0.37 1σ). This bias was correlated to the Th content of the sediment (Figure 2), and the slope of this correlation suggests that a false positive of +0.054 Bq kg^{-1} will be introduced per ppm Th. This bias is caused by the ^{228}Ac (^{232}Th series) emission at 478.3 keV (documented by Dalmasso *et al.*, 1987) which is unresolvable from the 477.6 keV ^{7}Be photon and needs to be corrected for during low-level measurements of ^{7}Be. Using a technique that is similar to the one thoroughly described in Landis *et al.* (2012), we measured a Th ore in our standard counting geometry to define a relationship between the ^{228}Ac emission at 478.3 keV and other ^{228}Ac lines (463 keV, 911 keV) that are easily quantified in samples. This relationship was used to project and subtract the ^{228}Ac interference from

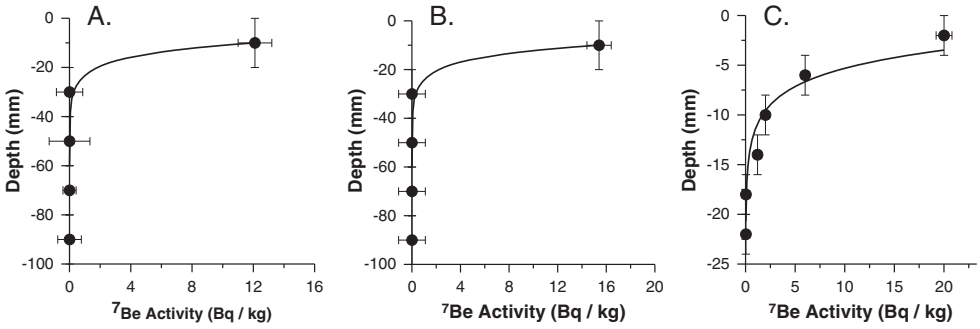

Figure 3. ^{7}Be activities in sediment profiles sampled from point bars on the West River (A,B) and from the gravel exposure experiment (C). Note the change in the y-axis scale in C

the net photopeak area at 477–478 keV in the unknown sample spectrums. To further evaluate the need and effectiveness of the ^{228}Ac interference correction, we repeatedly measured a sediment sample that had a moderate Th content (7 ppm) over four half-lives and calculated the decay rate (d^{-1}) using both the standard (uncorrected photopeak) and ^{228}Ac-corrected ^{7}Be photopeak. The uncorrected ^{7}Be measurements in this experiment 'decayed' from its initial ^{7}Be value (1.5 Bq kg^{-1}) at a much slower rate ($\lambda = 0.005$ d^{-1}) than corrected values ($\lambda = 0.01$ d^{-1}), indicating that the interference was substantial. Interference correction eliminates the positive bias and reduces the standard deviation of the repeated blank determinations, lowering our detection limits by nearly 50%. All sediment data we present here are corrected for the ^{228}Ac interference, and we calculate analytical detection limits to be 0.45 Bq kg^{-1} by multiplying the standard deviation of the interference-corrected blanks by the t-value for the 95% confidence interval ($n = 16$, Figure 2).

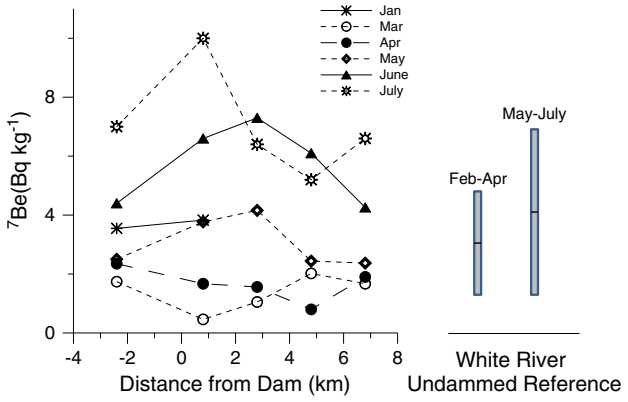

Figure 5. Monthly river sediment ^{7}Be (thalweg) in the impounded Black River (VT) in Early 2005. Sample location is given as relative distance downstream of the North Springfield Dam; the site with negative distance was sampled upstream of the dam-impacted reach. For comparison, the mean ^{7}Be and 1 standard deviation range is given for similarly collected samples from the unregulated White River (VT)

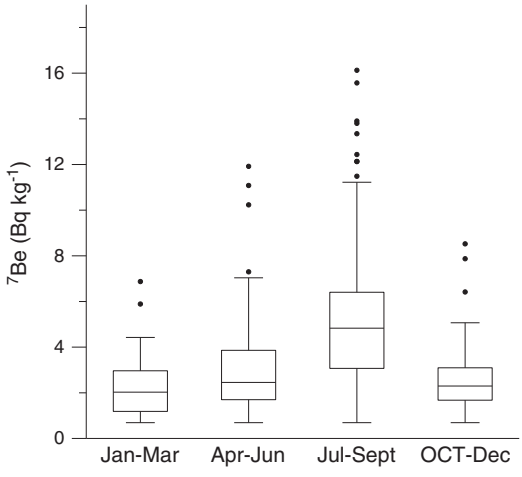

Figure 4. Box and whisker plot showing the seasonal distribution of ^{7}Be concentrations in transitional bedload sediment (<2 mm) collected from the upper few cm of the thalweg of rivers shown in Figure 1 from 2004 to 2008

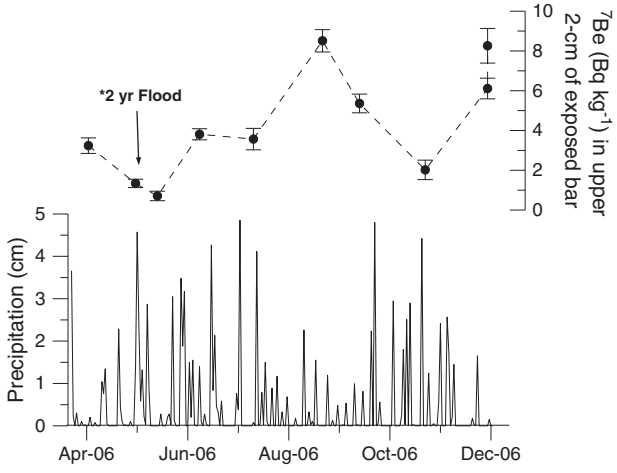

Figure 6. ^{7}Be concentration in the upper 2-cm layer of an exposed point bar measured approximately every month in Mink Brook. *Marks a rain-on-snow event in mid-May that generated a 2 year flow. Replicate samples were taken in December, and 1-sigma analytical error bars are given

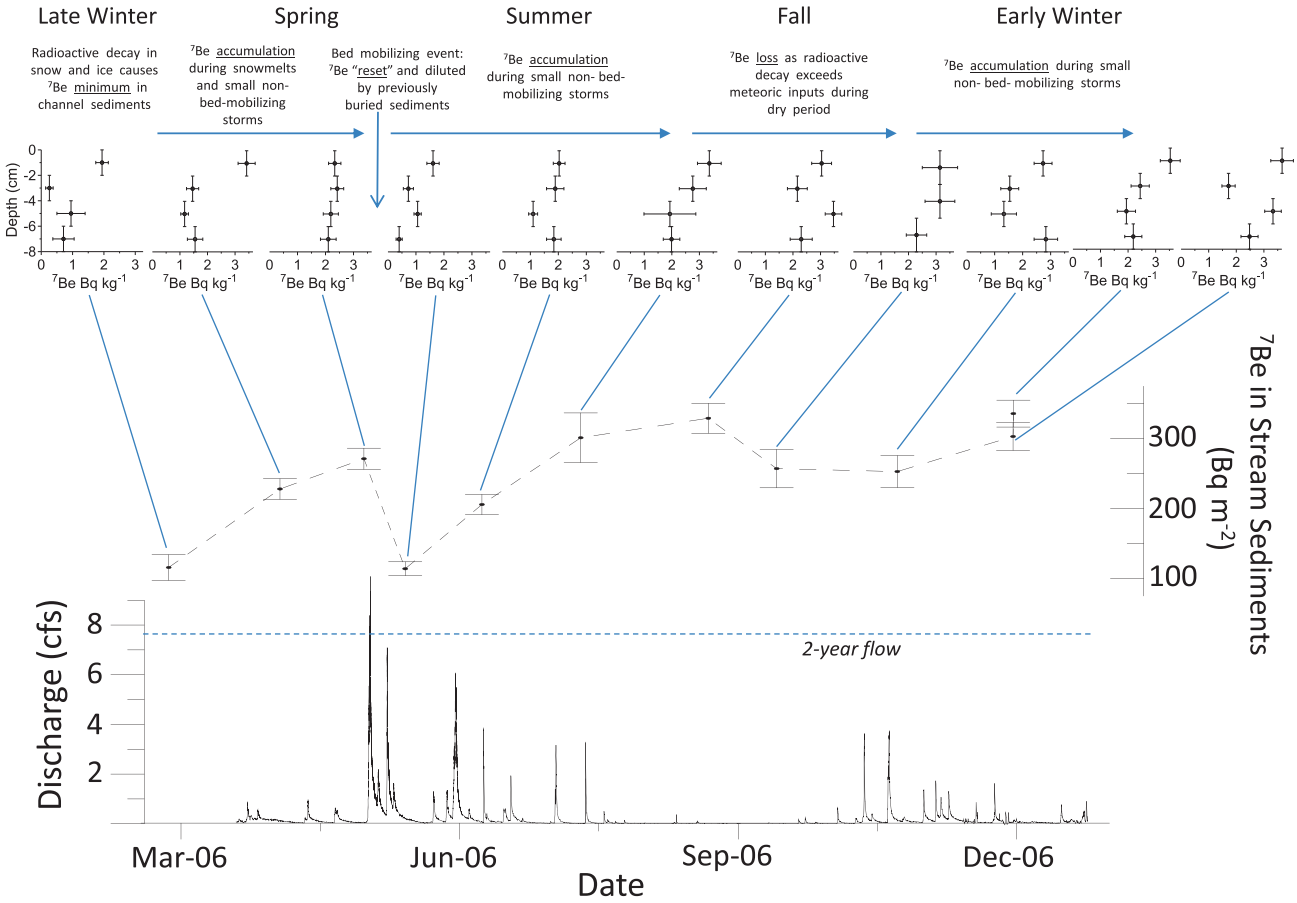

Figure 7. Seasonal variation of thalweg sediment ^{7}Be concentrations (upper) and inventories (middle) in Mink Brook in 2006. Corresponding maximum daily discharge data from a USGS gauging station are given (lower)

RESULTS AND DISCUSSION

Seasonal fluctuations in River Sediment ^{7}Be from atmospheric deposition

Our point bar profiles (Figures 3A-B) and gravel exposure experiment (Figure 3C) show how ^{7}Be is retained by the upper 10–20 mm of sediment that is exposed to rainfall. The inventory of ^{7}Be in the sediment in the exposure bin (Figure 3C) was $115 \pm 15\,\mathrm{Bq\,m^{-2}}$, which was equivalent to the inventory predicted by the ^{7}Be flux measurements ($98 \pm 12\,\mathrm{Bq\,m^{-2}}$). The two point bars had ^{7}Be inventories of 250 – 350 $\mathrm{Bq\,m^{-2}}$, all in the upper 2 cm. This inventory indicates near equilibrium with the atmosphere, as similar levels have been measured in stable forested soils nearby (Stankwitz *et al*., 2012). While an apparent exponential decline of ^{7}Be with depth has been reported for soils (Monaghan *et al*., 1983; Wallbrink and Murray, 1996), our data show that even unweathered sand and gravel scavenge ^{7}Be from meteoric waters rapidly. It follows that in fluvial environments, fresh material delivered to a channel, either by a flood, landslide, or bank collapse would accumulate ^{7}Be quantitatively over time, regardless of clay or organic matter content. The mechanism of Be adsorption onto quartz, feldspar, or mica grains may be explained by the formation of a direct bond between the Be^{2+} atom and oxygen on the surfaces or corners of minerals ('inner sphere adsorption'). Elements with large first hydrolysis constants ($K_h > 10^{-8}$) are usually stripped from waters by soils and sediments quickly in near-neutral to alkaline environments by this process (Nyffeler *et al*., 1984; Chen *et al*., 2006).

^{7}Be is readily detectable in stream sediments in the channel and on bars throughout the year (Figures 4–7). More than 90% of the sediment samples that we collected had detectable ^{7}Be ($n = 450$). In these rivers, point bars likely are a significant source of bedload sediment to the main channel during storm events (Salant *et al*., 2006). We show that ^{7}Be in point bar sediments changes by a factor of over four through the course of a year (Figure 6), which can be explained in part by seasonal fluctuations in meteoric ^{7}Be inputs and radioactive decay in the snowpack during winter months. Broadly speaking, in mid-latitude temperate zones, there is an accumulation of ^{7}Be in watershed soils and sediments during spring and summer because of stratosphere–troposphere mixing in the spring and increased convection and thunderstorms in

the early summer (Olsen *et al*., 1985; Baskaran *et al*., 1993; Zhu and Olsen, 2009). Dry periods during late summer and early fall cause a decrease in ^{7}Be surface inventories as decay exceeds meteoric inputs, and ^{7}Be amounts rise again in late fall as 'Nor'easters' and other large storms deliver intense rains to the region. Over the winter, frozen conditions cause a significant portion of the ^{7}Be to decay in the snowpack or ice rather than entering the sediments, so the point bar declines in ^{7}Be activity (Figure 6). This general seasonal trend is also apparent in sediments collected from the channels of the larger rivers (Figure 4) and in the impoundment-affected environments (Figure 5).

Submerged Mink Brook riffle sediment ^{7}Be fluctuates temporally similar to the point bar, with lowest ^{7}Be activities in late winter and highest activities in mid-summer (Figure 7), but the magnitude of change is considerably lower in the riffle than what we measured on the exposed point bar. At the end of winter, point bar sediment had ~1.5 Bq ^{7}Be kg^{-1}, and by mid-summer, concentrations rose to 8 Bq ^{7}Be kg^{-1}, whereas riffle sediment concentrations increased by only a factor of about two over the same time period (Figures 6,7). The median ^{7}Be concentration of all the larger river thalweg sediments also increased by a factor of approximately two from late winter to summer (Figure 4). Interestingly, the depth distribution of ^{7}Be in the riffle sediments is considerably different than what we measured in exposed sediments (Figure 3). In the stream channel, there is a package of sediments to at least 8-cm depth that maintains detectable ^{7}Be activities throughout the year during wet and dry periods (Figure 7). The depth distribution of ^{7}Be in this package would generally be better described as 'mixed' rather than the apparent exponential decay depth profile that we observed in point bars (Figure 3) and is more commonly reported for soils and sediments (Wallbrink and Murray, 1996; Blake *et al*., 1999). Our grain size measurements of the Mink Brook sediments indicate that riffle sediments (<2 mm) are well sorted sand (D_{50} mean $\pm 1\sigma = 0.91 \pm 0.09$ mm) with very small silt fractions (1–3% < 63 μm). Sediment surface areas were low but narrowly constrained to $1.3 \pm 0.2\ m^2\ g^{-1}$. We observed no significant correlation between ^{7}Be concentrations measured in Mink Brook sediments and D_{50} or percent silt ($r^2 < 0.02$; $n = 42$). This analysis indicates that in well-sorted sands, grain size effects on ^{7}Be scavenging and redistribution are not as substantial compared to other systems with larger fractions of silt and clay, in which radionuclide distributions can be dominated by transport of the fine-grained fraction (Bihari and Dezso, 2008). Thus, in moderate energy environments with a narrow range of grain sizes and low sediment surface areas, the use of ^{7}Be as a 'clock' may be simpler given that sediment sorting processes are less likely to impact concentrations and inventories.

^{7}Be in a sediment-supply limited environment below the NSD

The NSD on the Black River (Figure 1) maintains a relatively large storage reservoir (0.4 km^2) throughout the year, causing the majority of upstream-derived sediment to settle out. Detailed studies have documented armoring and incision below the dam (Salant *et al*., 2006; Magilligan *et al*., 2008), and it follows that sedimentary inputs of ^{7}Be would also be limited here. We therefore expected ^{7}Be depletion immediately below the NSD on the Black River because it is a supply limited environment with respect to recent sediment input. However, by comparing the downstream sediment samples to upstream sediment samples and the White River Reference Site, we found no consistent decrease in channel sediment ^{7}Be below the NSD (Figure 5). No consistent decrease was observed in the supply-limited environment below the TD on the West River, either (data not shown). Interestingly, we note that during May, June, and July, relatively high ^{7}Be activities were measured immediately below the NSD (Figure 5). This may be explained by limited removal of ^{7}Be from the reservoir's water column. In standing bodies of water, a lack of fines can limit the physical scavenging of ^{7}Be (Dominik *et al*., 1989), thereby allowing the $^{7}Be^{2+}$ ion persist. Given this scenario, impounded reservoirs could be a source of water enriched in $^{7}Be^{2+}$ which would then be scavenged by bed sediments downstream of the dam, but more sampling of outflow waters would be needed to assess this mechanism.

It is clear that ^{7}Be concentrations can be affected by sediment supply and transport dynamics below dams depending on how the impoundment is managed. Salant *et al*. (2007) found that thalweg ^{7}Be was reduced below the UVD on the Ompompanoosic River in Vermont (Figure 1) after the Spring flood release caused a pulse of sediment depleted in ^{7}Be to mix into the channel sediments. Because the UVD does not maintain a reservoir during the summer and the gates are opened completely in Spring, aggradation occurs downstream as erosion is unable to keep pace with the sediment supply (Magilligan *et al*., 2008). Large pulses of sediment released by the dam cause a dilution of the ^{7}Be signal in the tailwater sediments, especially in the Spring (Salant *et al*., 2007). While dams can create both supply-limited and transport-limited conditions, the seasonal ^{7}Be variations that we observe in our broader dataset (Figure 4) are apparent in both regulated and unregulated rivers (Figure 5), and we conclude that fresh sediment supply does not necessarily limit ^{7}Be activities in channels.

Seasonal fluctuations in channel ^{7}Be from sediment exchange during large hydrologic events

Because of its short half-life and particle-reactive behaviour, ^{7}Be is a commonly used indicator of 'recent'

sediment; that is, recently eroded topsoil (Bonniwell *et al*., 1999; Matisoff *et al*., 2002), recently mobilized bar or bank sediments (Fisher *et al*., 2010), or sediments that have recently been re-suspended in the water column, scavenged ^{7}Be, and subsequently deposited as a flood package (Feng *et al*., 1999; Fitzgerald *et al*., 2001). If we extend this framework to channel environments, it follows that thalweg sediments that are locked in place for a period of a few months when there were no bed-mobilizing events would decay and have low ^{7}Be activities. Our precipitation, hydrograph, and ^{7}Be data from Mink Brook actually show that the opposite scenario can unfold. During late winter and early spring of 2006, thalweg sediment accumulated ^{7}Be during a time period of frequent rain and 'rain on snow' events that were too small to mobilize the bed (Figure 7). The upper 8 cm of channel sediment accumulated a ^{7}Be inventory of ~300 Bq m^{-2}, which indicates near-equilibrium with the atmosphere by the end of April (Figure 7). Then, in May, a large hydrologic event (>2 year flow) caused ^{7}Be concentrations to plummet by 60%, resetting the channel ^{7}Be to well-below levels that could be supported by the atmosphere. While we anticipated that freshly deposited sediments would have high ^{7}Be concentrations, it seems more likely that this event deposited sediments that were depleted in ^{7}Be, possibly because the material was recently eroded from considerable (>8 cm) depth. One could certainly envision the opposite scenario, where a different sediment source could produce a 'new' deposit with high ^{7}Be.

Another interesting aspect of our data is that sediments that are effectively stationary in the thalweg can maintain near-steady ^{7}Be activities over a timescale of a half-life or so- in the absence of significant fresh sediment inputs or large rain events. The summer of 2006 was relatively dry, with a few thunderstorms that didn't impact the hydrograph significantly, but despite the lack of substantial sediment inputs or exchange, riffle sediment ^{7}Be activity increased from the end of June through the end of August, again approaching equilibrium with the atmosphere (Figure 7). Small rainfall events can have high concentrations of ^{7}Be (in Bq ^{7}Be per l of rainfall, Olsen *et al*., 1985; Papastefanou and Ioannidou, 1991; Baskaran, 1995) but have a negligible effect on the hydrograph. During these events, streamwater will directly receive ^{7}Be, which could get stripped from the water as it flows through the alluvial bed. Alternatively, light rain events could mobilize colloids or fine particulate matter from the near-shore edges of bars or banks, and this ^{7}Be-enriched material could get sequestered by channel sediment (Hamm *et al*., 2009; Gartner *et al*., 2012). While in estuarine and coastal shelf environments, a layer of sediment enriched in ^{7}Be is commonly interpreted as a newly deposited package (Larsen and Cutshall, 1981; Sommerfield *et al*., 1999), in river environments this may not be the case. Channel sediments can receive inputs of ^{7}Be even when pulses of 'new' sediment input are limited (Figure 5, Figure 7). Between the end of August and the beginning of October, ^{7}Be concentrations in thalweg sediments declined, because rainfall was virtually absent during this period, allowing radioactive decay to outpace ^{7}Be inputs (Figures 6, 7).

^{7}Be dynamics in fluvial environments and potential applications

Cosmogenic ^{7}Be is delivered to watersheds primarily via wet precipitation, but only a fraction of rainfall events mobilize the bed and result in significant sediment exchange in well-sorted sandy environments. It is clear that nearly every precipitation event will introduce ^{7}Be to hillslopes and exposed bars, and these exposed units will develop strong concentration–depth gradients of ^{7}Be. We show that channel sediments maintain or develop increasing ^{7}Be content in a relatively mixed layer to a depth of at least 8 cm even when naturally or artificially low–flow conditions prevent fresh sedimentary inputs or exchange. It is probable that during small to intermediate storms that do not generate much sediment, ^{7}Be is delivered to the water column, perhaps directly as the $^{7}Be^{2+}$ ion, and as this water mixes rapidly with the bed during low flow, there is exchange between the channel sediment and the $^{7}Be^{2+}$ ion in the hyporheic zone. It is also possible that the $^{7}Be^{2+}$ is adsorbed to very fine colloids of aluminum, iron, and/or silica which are common in granitic landscapes (Farmer and Fraser, 1982). Because even coarse-grained soils and sediment bars appear to be very effective at scavenging ^{7}Be from meteoric waters (Figure 3), the simplest explanation is that rain falling directly on the stream delivers ^{7}Be as an ion or an adsorbed phase to the water column which then mixes with the stream bed. Total inventories in the 0–8 cm layer of sediment in the thalweg approached 300 Bq m^{-2} several times throughout the year (Figure 7), which is very near the equilibrium levels that we would expect from atmospheric fallout on a stable surface in our region. If ^{7}Be essentially builds up in channel sediments from hyporheic exchange (Gartner *et al*., 2012), then the activity of ^{7}Be in the sediments will be a function of flow velocity and the timescale of geomorphic stability. Consequently, sediments that remain in place in a high velocity zone of hyporheic flow for a short duration of time might accumulate a similar level of ^{7}Be as sediments in place in a low velocity zone for a longer period of time.

The conceptual framework that sediments locked in the channel can accumulate ^{7}Be over time and sediment-

starved channels can have high ^{7}Be diverges somewhat from the framework applied in estuarine and coastal systems- where ^{7}Be is used to trace newly deposited packages of sediment (Sommerfield *et al*., 1999). Moreover, if ^{7}Be concentrations and inventories scale with the timescale of bed stability and/or flow rates, then this tracer may be used to link aquatic ecosystem health and organism behaviour to sediment dynamics. Svendsen *et al*. (2009) showed a significant relationship between sediment ^{7}Be (normalized to ^{210}Pb) and Trichoptera abundance measured in the West River in August of 2006. Trichoptera larva construct nets that are fixed to the substrate to catch detritus and thus seek sections of the riverbed that are stable in moderate to high flows. If ^{7}Be concentrations in river sands scale with the amount of water that the sediment has been in contact with, ^{7}Be and Trichoptera would be correlated, as ^{7}Be is essentially 'tagging' environments that are favourable to filter feeders. Svendsen *et al*. (2009) also noted that the activity of the mainstem river sediment increases immediately downstream of the largest tributaries during the summer sampling period. While it was concluded that the tributaries were a source of 'fresh' sediments that were recently tagged with ^{7}Be, it is just as likely that submerged sediments immediately downstream of large tributaries accumulate more ^{7}Be because the flux of water passing through the sediments increases at these junctions. We note that below the NSD, sediment ^{7}Be concentrations were actually highest immediately downstream of the dam during June and July (Figure 5). Detailed studies of ^{7}Be in lakes have shown that the residence time of the $^{7}Be^{2+}$ ion in still waters is on the order of months, because the low availability of particles can limit adsorption (Dominik *et al*., 1989; Steinmann *et al*., 1999). The reservoir upstream of the NSD may accumulate ^{7}Be in the water column from rainfall, perhaps as a dissolved ion, but when the water passes through the dam and mixes with the bed in the free-flowing section, the ^{7}Be may be scavenged from the water by the stationary sands in the channel.

Our observations that large hydrologic events can deplete channel ^{7}Be, and that during periods of moderate rainfall, ^{7}Be activities to depths of 8 cm may scale with bed stability (Figure 7) are relevant to the specific riffle and unobstructed channel environments that we sampled here. In depositional environments in the Ducktrap River in Maine, fresh sediment inputs appeared to introduce a steady supply of ^{7}Be to the surface of the bed (Fisher *et al*., 2010). Thus, the low-flow hyporheic replenishment of ^{7}Be that we observed may only be applicable to riffles and thalweg sediments rather than depositional environments downstream of wood or boulders or embedded transport-limited reaches. Our observations here and previous studies (Salant *et al*., 2007) indicate that large pulses of 'new' sediment can have a dilution effect on ^{7}Be activities in fluvial systems (Figure 7). Thus, measuring ^{7}Be before and after events may be the most useful approach for quantifying the spatial extent of sediment exchange and characterizing sediment source areas.

CONCLUSIONS

When rainfall is intercepted by sediment exposed within fluvial channels, ^{7}Be is quantitatively retained by grains in the upper one to two cm of the profile. The adsorption of ^{7}Be does not require clays or organic matter, and in well-sorted sandy environments, ^{7}Be concentrations do not appear to be controlled by grain size sorting effects. By studying supply-limited conditions below dams and in Mink Brook during dry months, we show that channels can maintain ^{7}Be sediment activities in the absence of large fresh sedimentary inputs. The ^{7}Be content of river sediment is controlled by both seasonal variations in meteoric inputs and large-scale sediment exchange processes. In the riffles and in the thalweg, a layer of sediment at least 8 cm thick is approximately mixed with respect to ^{7}Be. This layer of sediment can have maintained or increasing ^{7}Be activity with time in the absence of significant hydrologic events and large-scale sediment exchange if small rainstorms periodically deliver ^{7}Be to the water column and surrounding parts of the watershed. After an extreme flood, we observed a decrease in thalweg ^{7}Be, as sediment depleted in ^{7}Be replaced sediment that had high ^{7}Be activities. Our data suggest that ^{7}Be concentrations in submerged sediments may reflect timescale of stability and/or average flow velocity, but it is certainly possible that a large bed-mobilizing event could result in the deposition of ^{7}Be enriched material if the sediment source area was high in initial ^{7}Be. When sediment ^{7}Be concentrations are considered within the context of seasonal fallout variability and timescales of bed stability, the nuclide can be a powerful tracer of mass transport and exchange in fluvial environments.

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