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18 Erosion rates and sediment flux within the Potomac River basin 19 quantified over millennial timescales using beryllium isotopes

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31 ABSTRACT

32 Beryllium isotopes measured in detrital river sediment are often used to estimate rates of
33 landscape change at a basin scale, but results from different beryllium isotope systems have
34 rarely been compared. Here, we report measurements of *in situ* and meteoric ¹⁰Be_i (¹⁰Be_i and
35 ¹⁰Be_m, respectively) along with measurements of reactive and mineral phases of ⁹Be (⁹Be_{react} and
36 ⁹Be_{min}, respectively) to infer long-term rates of landscape change in the Potomac River basin,
37 North America. Using these data, we compare directly results from the two different ¹⁰Be isotope
38 systems and contextualize modern sediment flux from the Potomac River basin to Chesapeake
39 Bay.

40 Sixty-two measurements of ¹⁰Be_i in river sand show that the Potomac River basin is
41 eroding on average at $29.6 \pm 14.1 \text{ Mg km}^{-2} \text{ yr}^{-1}$ ($11 \pm 5.2 \text{ m m.y.}^{-1}$ assuming a rock density of
42 $2,700 \text{ kg m}^{-3}$) – a rate consistent with other estimates in the mid-Atlantic region. ¹⁰Be_i erosion
43 rates correlate with basin latitude, suggesting that periglacial weathering increased with
44 proximity to the former Laurentide Ice Sheet margin. Considering the ¹⁰Be_i-derived erosion rate
45 as a sediment flux over millennia, rates of sediment delivery from the Potomac River to

46 Chesapeake Bay are ~10x lower than contemporary sediment yields implying modern land-use
 47 practices have accelerated erosion and sediment transport over background rates. However, $^{10}\text{Be}_i$
 48 erosion rate data suggest that regulatory benchmark levels used to manage sediment export from
 49 the Potomac River basin to Chesapeake Bay are set appropriately to reduce sedimentation and
 50 restore the Bay's ecological health.

51 The mean of $56\text{ }^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ -derived denudation rates ($40.0 \pm 21.7\text{ Mg km}^{-2}\text{ yr}^{-1}$) is
 52 higher than, but statistically indistinguishable from, the mean $^{10}\text{Be}_i$ erosion rate ($29.6 \pm 14.1\text{ Mg km}^{-2}\text{ yr}^{-1}$; $p = 0.003$). However, when considered basin by basin, $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ -determined
 53 denudation rates are only weakly correlated ($R^2=0.208$; $p < 0.001$) with sediment fluxes
 54 determined from the well-established and widely used $^{10}\text{Be}_i$ technique. This suggests that the
 55 $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ technique may not reflect the same geomorphic processes as $^{10}\text{Be}_i$ technique, or that
 56 the $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ technique operates over different time and/or depth scales. Erosion indices (EIs,
 57 *sensu* Brown et al., 1988) derived from $^{10}\text{Be}_m$ measurements and contemporary sediment yield
 58 data range from 0.07 to 1.24; 75% of basins sampled have EIs that are similar to or greater than
 59 1, suggesting that $^{10}\text{Be}_m$ is being retained and sediment is being stored within the Potomac River
 60 basin. The Appalachian Plateau is the only physiographic province where sediment export
 61 dominates, likely as the result of on-going relief growth in catchments draining the Appalachian
 62 Mountain divide. $^{10}\text{Be}_m$ concentrations measured in the 150 k.y. Hybla Valley sediment core,
 63 taken from the lower Potomac River basin, suggest that $^{10}\text{Be}_m$ and sediment is preferentially
 64 stored in the catchment when vegetation proxies for climate change suggest warmer conditions
 65 prevail. $^{10}\text{Be}_m$ and sediment are exported when vegetation proxies for climate suggest conditions
 66 are colder, perhaps a reflection of periglacial activity or changes in storm frequency and/or
 67 magnitude over glacial/interglacial cycles.

68
 69
 70 **INTRODUCTION**
 71 Understanding the effects that human land-use practices have on landscapes requires
 72 knowledge of background (geological) rates of sediment erosion and denudation, transport, and
 73 deposition (Hooke et al., 2012; Pelletier et al., 2015). Despite several decades of intensive data
 74 collection (Judson, 1968; McLennan, 1993; Milliman and Syvitski, 1992; Portenga and Bierman,
 75 2011), there remain many landscapes for which there is little quantitative information about
 76 natural or background rates of landscape change (e.g., Arkle et al., 2017; Jonell et al., 2016;
 77 Mandal et al., 2015; Reusser et al., 2015, Struth et al., 2017). The Potomac River basin along the
 78 United States' east coast is a landscape where large volumes of sediment deposition in
 79 Chesapeake Bay are known to have resulted from widespread erosion associated with intensive
 80 European-American land-use practices in the 1700–1800s (Fig. 1; Brush, 2009; Costa, 1975;
 81 Kirby, 2004; Saenger et al., 2008). However, the effect of this human-induced land-use change
 82 on erosion and sediment transport rates remains only loosely constrained because background
 83 rates and spatial patterns of pre-disturbance landscape change are largely unknown.

84 Since the 1980s, the use of isotopic tracers, specifically the cosmogenic isotope ^{10}Be , has
 85 greatly increased our knowledge of the rate at which Earth surface processes operate (e.g., Graly
 86 et al., 2010; Harel et al., 2016; Portenga and Bierman, 2011; Willenbring and von Blanckenburg,
 87 2010). Both the ^{10}Be produced by cosmic-ray interactions in the atmosphere (meteoric, $^{10}\text{Be}_m$)
 88 and that created by interactions in mineral grains at Earth's surface (*in situ*, $^{10}\text{Be}_i$) have been
 89 measured and used to infer process rates and trace sediment across the landscape (e.g., Brown et
 90 al., 1998; Helz and Valette-Silver, 1992; Kirchner et al., 2001; Ouimet et al., 2009; Portenga et
 91 al., 2017; Reusser and Bierman, 2010; Schaller et al., 2001; You et al., 1988). Recently, the

92 stable isotope of beryllium, ^{9}Be , has been measured in soil and sediment, and its abundance has
93 been used to normalize measured activities of $^{10}\text{Be}_m$ produced in the atmosphere and
94 incorporated in soil and sediment grain coatings (Dannhaus et al., 2018; Rahaman et al., 2017;
95 von Blanckenburg et al., 2012; Wittmann et al., 2015). This ^{9}Be normalization approach was
96 developed, in part, for the purpose of calculating denudation rates (total mass loss per unit area
97 over time) at the basin scale in areas where quartz was not available for the now well-established
98 $^{10}\text{Be}_i$ erosion rate technique (Dannhaus et al., 2018; von Blanckenburg et al., 2012), which in
99 most settings gives values for erosion (the mass of sediment removed per unit area over time;
100 Lal, 1991).

101 In this paper, we report measurements of $^{10}\text{Be}_i$ and $^{10}\text{Be}_m$ in conjunction with
102 measurements of reactive ^{9}Be ($^{9}\text{Be}_{\text{reac}}$, in this case, acid-extractable) coating sediment grains and
103 unweathered ^{9}Be ($^{9}\text{Be}_{\text{min}}$) contained within the mineral grains of bulk sediment. We use the $^{10}\text{Be}_i$
104 data to investigate spatial and temporal patterns of erosion, and by inference sediment flux, in the
105 Potomac River basin in order to gain an understanding of the rates at which mass was delivered
106 to Chesapeake Bay prior to the impacts of European-Americans settling the region. Together,
107 these data allow us to compare rates of landscape change (erosion and denudation) calculated
108 from $^{10}\text{Be}_i$ and $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ ratios (cf. von Blanckenburg et al., 2012) measured in the same
109 samples. Contemporary sediment yield data are available for ten basins in the Potomac River
110 watershed (Gellis et al., 2004); we compare these data to long-term erosion rates inferred from
111 $^{10}\text{Be}_i$. We also employ the method developed and tested by Brown et al. (1988) using $^{10}\text{Be}_m$ to
112 calculate erosion indices (EIs) for ten basins draining different physiographic provinces within
113 the watershed. This method assesses the isotopic balance between $^{10}\text{Be}_m$ entering and leaving a
114 basin; because $^{10}\text{Be}_m$ is adhered to sediment grain coatings, the EI serves as a proxy for the
115 amount of sediment being retained within or exported from a basin. To provide a long-term
116 context for the contemporary isotopic data, we also measured $^{10}\text{Be}_m$ in 13 samples spanning the
117 last 150 k.y., isolated from a sediment core collected in Hybla Valley, an abandoned meander of
118 the lower Potomac River (Litwin et al., 2013).

119 In addition to providing specific information about the Potomac River basin, our data
120 provide the first independent large-scale comparison between the two different ^{10}Be isotopic
121 systems used to understand rates of basin-scale landscape change from detrital, fluvial sediment.
122 We use the entire data set to address a variety of outstanding questions: How well-correlated are
123 long-term denudation rates estimated using $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ and erosion rates quantified using
124 $^{10}\text{Be}_i$? Are contemporary, short-term sediment yields similar to or different from long-term rates
125 of landscape change inferred from Be isotopes? What roles do topography, landscape
126 physiography, and human land use play in changing the landscape of the Potomac River basin?
127

128 Field Area

129 The Potomac River drains \sim 38,000 km 2 of the central Appalachian Mountains and is a
130 major source of sediment to Chesapeake Bay, the largest estuary in the United States (Gellis et
131 al., 2004; Fig. 1). Despite the importance of the Bay and its ecosystems to the ecology and
132 economy of the mid-Atlantic region, little is known about the long-term rates of sediment export
133 from the Potomac River basin to the Bay. Erosion rates of summit ridgelines throughout the
134 central Appalachian Mountains, inferred from measured concentrations of $^{10}\text{Be}_i$ in bedrock
135 outcrops, are on the order of \sim 10 m m.y. $^{-1}$ (Duxbury et al., 2015; Hancock et al., 2015; Portenga
136 et al., 2013). However, outcrops exist on ridgelines because they erode more slowly than the
137 drainage basins they divide; inferring erosion rates or sediment generation rates over large areas

138 (drainage basin scale) from outcrop data alone is not reasonable because of this bias.

139 Sediment in the Potomac River basin is derived from five physiographic provinces
140 (Fig. 2). These provinces are delineated by significant changes in bedrock lithology, structural
141 features, and topography. From west to east across the Potomac River basin, the physiographic
142 provinces include: (1) the Appalachian Plateau, the western, mostly undeformed part of the
143 foreland, which is underlain by Upper Paleozoic clastic and carbonate rocks; (2) the Valley and
144 Ridge Province, which consists of Lower Paleozoic clastic and carbonate rock in the eastern,
145 deformed part of the foreland, where they are folded into elongate synclines and anticlines; (3)
146 the Blue Ridge, which is a mix of quartzite and volcanic rocks, widely recognized for providing
147 the steep topography that makes up Shenandoah National Park; (4) the Piedmont region, which
148 consists of rolling hills covered by mature soils and which is underlain by deeply weathered and
149 heavily deformed Proterozoic gneiss (which formed the core of the Alleghenian orogeny); and
150 (5) the Coastal Plain, which sits east of the Fall Line and consists mostly of Cretaceous and
151 younger reworked fluvial, estuarine, and nearshore and marine sediments. Quartz-bearing rocks
152 underlie all of these provinces.

153 Although the Potomac River basin was never glaciated, extensive periglacial weathering
154 occurred at high elevations throughout the central Appalachians and is thought to have increased
155 the amount of unconsolidated material available for delivery to offshore basins (Clark and
156 Ciolkosz, 1988; Denn et al., 2018; Poag and Sevon, 1989; Whittecar and Ryter, 1992).
157 Additionally, long-term flexural hydro-isostatic uplift of the land surface in and near the
158 Potomac basin, which was the result of continued offshore sediment loading (Pazzaglia and
159 Gardner, 1994), punctuated by higher-frequency glacial-isostatic adjustments (DeJong et al.,
160 2015; Peltier, 1996), drove complex histories of land surface change and possibly caused higher
161 rates of river incision near the Fall Line at Great Falls during times in the past (Bierman, 2015;
162 Reusser et al., 2004).

163 Widespread land-use change in the Chesapeake Bay watershed began in the 1700s when
164 European colonists, drawn by the fertile soils of the Appalachian Piedmont region and the vast
165 oyster stocks in the Bay, moved into the region (Brown et al., 1988; Brush, 2009; Colman and
166 Bratton, 2003; Cooper and Brush, 1993; Costa, 1975; Gellis et al., 2004; Kirby, 2004; Langland
167 and Cronin, 2003; Montgomery, 2007; Saenger et al., 2008; Wolman, 1967). Up to 80% of land
168 within the Chesapeake Bay watershed was deforested and used for agriculture by the late 1800s,
169 which caused rapid erosion and delivery of topsoil, nutrients, and charcoal to the Bay at levels
170 much higher than natural background (Brush, 2009; Cooper and Brush, 1993; Valette-Silver et
171 al., 1986).

172 Only a few low dams have been constructed across the Potomac River (Gerhart, 1991),
173 and therefore most of Potomac River's bedload is transported directly to Chesapeake Bay;
174 however, milldams on small Potomac River tributaries effectively trapped sediment eroded after
175 deforestation by European settlers (Walter and Merritts, 2008). These milldams have fallen into
176 disrepair, and once-trapped sediment is now being released into the larger Potomac River basin.
177 For the period of 1952–2001, the Potomac River was the largest contributor of sediment to the
178 Bay, supplying ~44% of the total sediment flux (Gellis et al., 2004); this amount exceeds
179 sediment yields from the larger, previously glaciated Susquehanna River basin to the north (Ives,
180 1978). However, Susquehanna River is heavily dammed, which likely leads to sediment
181 retention and decreases sediment transport to the Bay, except during large floods, when sediment
182 is scoured from the beds of reservoirs and transported downstream (Langland and Cronin, 2003;
183 Langland and Hainly, 1997).

184 Human impacts since the 1700s and continuing until today have been particularly severe
185 in and near the heavily populated urban areas of Baltimore and Washington, DC, where
186 development has disturbed much of the land and increased sediment delivery to Chesapeake Bay
187 (Brush, 2009; Dauer et al., 2000; Gellis et al., 2017; Wolman, 1967). Restoration of the Bay's
188 ecological health depends, in part, on reducing sediment delivery from tributary rivers to pre-
189 disturbance levels (Hassett et al., 2005; Langland and Cronin, 2003). Doing so requires
190 knowledge of the spatial patterns and background rates of erosion and denudation in landscapes
191 upstream from the Bay. This is a motivation for our study.

192 Isotopic Tracers

193 Cosmogenic ^{10}Be is produced as cosmic rays interact with O and N atoms both in rock
194 and sediment at Earth's surface ($^{10}\text{Be}_i$, Lal and Peters, 1962) and in the atmosphere ($^{10}\text{Be}_m$;
195 Brown, 1988). $^{10}\text{Be}_m$ subsequently falls to Earth's surface with precipitation or as dry fallout and
196 then adheres to the surfaces of mineral grains in soil and sediment profiles, where it is
197 incorporated into pedogenic grain coatings (Brown et al., 1981; Dixon et al., 2017; Graly et al.,
198 2010; Pavich et al., 1985; Willenbring and von Blanckenburg, 2010). In this study, we measured
199 isotopic concentrations of $^{10}\text{Be}_i$ and $^{10}\text{Be}_m$ in sand-sized detrital fluvial sediment, using the data
200 as indicators of sediment sourcing, rates of landscape change, and spatial patterns of landscape
201 behavior.

202 The isotope $^{10}\text{Be}_i$ is produced in the mineral lattices of rock and sediment at Earth's
203 surface, and measuring its abundance in quartz has become widely accepted as a way of deriving
204 long-term erosion rates of rock outcrops and drainage basins, as well as rates of soil formation
205 (e.g., Bierman and Steig, 1996; Brown et al., 1995; Granger et al., 1996; Heimsath et al., 2006;
206 Nishiizumi et al., 1986; and numerous other papers as summarized in Harel et al., 2016; Portenga
207 and Bierman, 2011; Willenbring et al., 2013). After being produced in rock and soil on slopes,
208 the regolith and the $^{10}\text{Be}_i$ that it contains are then eroded. This eroded sediment is eventually
209 delivered to rivers, where it is mixed such that $^{10}\text{Be}_i$ concentrations measured in alluvium
210 collected at any point along a stream can be used to derive a long-term erosion rate that averages
211 over the upstream contributing area. The duration over which erosion is integrated is determined
212 by the time it takes to erode through one cosmic-ray attenuation depth, ~60 cm of rock, and thus
213 isotope concentration is inversely related to landscape stability (Lal, 1991) and biased toward the
214 surface and recent erosion history. In slowly eroding, areas, such as the Appalachian Mountains,
215 which occupy much of the Potomac River basin, $^{10}\text{Be}_i$ erosion rates integrate over 10^4 to 10^5 yr
216 (e.g., Duxbury et al., 2015; Hancock and Kirwan, 2007; Linari et al., 2016; Matmon et al., 2003;
217 Portenga et al., 2013; Reusser et al., 2015).

218 Slow erosion rates allow time for $^{10}\text{Be}_m$ to accumulate and be retained on sediment grain
219 coatings during pedogenesis and before regolith is eroded and transported (Barg et al., 1997;
220 Greene, 2016; Wittmann et al., 2015). If grain coatings are stable and there is negligible loss of
221 $^{10}\text{Be}_m$ from soils in the dissolved phase, then concentrations of $^{10}\text{Be}_m$ would be correlated with
222 concentrations of $^{10}\text{Be}_i$ in sediment and thus anticorrelated with $^{10}\text{Be}_i$ erosion rates and other
223 metrics of $^{10}\text{Be}_i$ -derived sediment fluxes. Such relationships have been observed in the whole of
224 the Amazon River basin (von Blanckenburg et al., 2012). The isotope $^{10}\text{Be}_m$ has also been used
225 as a sediment tracer in a variety of fluvial systems (Brown et al., 1988; Helz and Valette-Silver,
226 1992; Portenga et al., 2017; Reusser and Bierman, 2010; Valette-Silver et al., 1986; van Geen et
227 al., 1999). However, identifying sediment sources in basins as large as the Potomac River basin
228 is challenging.

229 Questions about $^{10}\text{Be}_m$ stability after incorporation into sediment grain coatings, and thus

230 geomorphological interpretations based on measurements of $^{10}\text{Be}_m$ concentration in these
 231 coatings, have motivated the search for a method to normalize $^{10}\text{Be}_m$ concentrations using
 232 another isotope with similar chemical behavior (Bacon et al., 2012; Wittmann et al., 2012, 2015).
 233 Once regolith is eroded from rock outcrops and subcrops, most native ^9Be ($^9\text{Be}_{\text{parent}}$) remains
 234 within silicate mineral crystal lattices ($^9\text{Be}_{\text{min}}$); however, chemical weathering releases some ^9Be
 235 to soil solutions and groundwater (von Blanckenburg et al., 2012). The ^9Be that is incorporated
 236 into sediment grain coatings is termed the reactive phase ($^9\text{Be}_{\text{reac}}$), and the dissolved portion is
 237 termed the dissolved phase ($^9\text{Be}_{\text{diss}}$; von Blanckenburg et al., 2012, Wittmann et al., 2015). ^9Be is
 238 thought to adhere to mineral grains via the same mechanisms as $^{10}\text{Be}_m$ (Barg et al., 1997; von
 239 Blanckenburg et al., 2012); thus, the $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ ratio is assumed to be locked at the time of
 240 coating deposition and can be considered a closed system (Graly et al., 2010). Based on this
 241 closed-system model, the $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ ratio of authigenic minerals or sediment grain coatings
 242 has been proposed as an independent measure of rates of landscape change (von Blanckenburg et
 243 al., 2012). In the laboratory, this weathering-sourced $^9\text{Be}_{\text{reac}}$ can be removed from sediment
 244 grains using a variety of methods that have been shown to remove little, if any, mineral lattice
 245 ^9Be (i.e., $^9\text{Be}_{\text{min}}$) incorporated in silicate minerals (Greene, 2016; Wittmann et al., 2012).

246 Erosion and denudation are often confused and/or equated but their definitions are
 247 important and distinct in the context of ^{10}Be production and/or delivery and retention on
 248 landscapes. Here, we define erosion measured using $^{10}\text{Be}_i$ as the mass of solid material lost from
 249 an area of Earth's surface over time (a rate, $\text{M L}^{-2} \text{T}^{-1}$). Assuming a density (M L^{-3}), erosion can
 250 be reported in units of L T^{-1} . Erosion rates, as described below, can be derived from both $^{10}\text{Be}_i$
 251 and $^{10}\text{Be}_m$ and, assuming steady state, one can equate the mass of physical material supplied by
 252 erosion over time to the sediment flux out of a basin; in other words, all eroded sediment ends up
 253 in the river eventually with no net change in long-term sediment storage. In contrast, denudation
 254 is the total physical and chemical mass lost per unit area over a given duration of time, again in
 255 units of $\text{M L}^{-2} \text{T}^{-1}$. In this study, denudation rates are inferred from $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ ratios. By
 256 definition, denudation includes both erosion and mass lost by dissolution and therefore
 257 denudation rates cannot be lower than erosion rates or inferred long-term sediment fluxes.

258 Depending on the geomorphic setting, $^{10}\text{Be}_m$ and $^{10}\text{Be}_i$ may integrate mass loss rates over
 259 different depths. $^{10}\text{Be}_i$ is most sensitive to physical mass loss in the uppermost few meters of the
 260 soil/regolith system – the depth of $^{10}\text{Be}_i$ production through fast neutron penetration and
 261 spallation. In contrast, the $^{10}\text{Be}_m$ system begins recording information about the cumulative
 262 physical and dissolved mass loss at the weathering front, which in some settings can be much
 263 deeper than the penetration depth of most cosmic rays. For example, in the Potomac River basin,
 264 the weathering front is within the cosmic ray penetration depth over much of the Appalachian
 265 Plateau, Valley and Ridge, and the Blue Ridge, where soils are shallow on steep hillslopes.
 266 However, in the Piedmont, meters of saprolite can overly the weathering front (Pavich, 1989,
 267 1990); thus, mass in the Piedmont is lost by solution below the penetration depth of most cosmic
 268 rays. As a result of mass loss by solution below the penetration depth of cosmic rays, there exists
 269 the possibility that quartz grains remaining in soil profiles reside on the landscape longer than the
 270 residence time of bulk, non-quartz material (Riebe et al., 2001). In this case, measurements of
 271 $^{10}\text{Be}_i$ result in erosion rates that are less than the rate of total denudation, as recorded by the
 272 $^{10}\text{Be}_m$ system, because $^{10}\text{Be}_i$ does not directly track mass loss by solution at depth. Such thinking
 273 is not relevant to the Coastal Plain, which is made up predominantly of reworked quartz-rich
 274 sand and gravel with few primary minerals left to weather. It is important, therefore, to make the
 275 distinction between $^{10}\text{Be}_i$ erosion rates, which refer primarily to the physical mass loss from

276 landscapes within the $^{10}\text{Be}_i$ nuclide production depth and $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates, which
277 model total mass loss from the entire depth of regolith.

278 METHODS

279 We collected stream sediment from 70 sub-basins within the Potomac River watershed
280 (Fig. 2; Table DR1¹) for analysis of $^{10}\text{Be}_i$, $^{10}\text{Be}_m$, $^{9}\text{Be}_{\text{reac}}$, and $^{9}\text{Be}_{\text{min}}$. Of these, 10 samples were
281 collected at locations with present or former U.S. Geological Survey (USGS) gauging stations
282 and associated documented long-term annual sediment yields (Gellis et al., 2004). Most samples
283 were collected from small basins (5–100 km²) in each of the five physiographic provinces
284 drained by Potomac River. During sample collection, we observed minimal signs of major
285 human disturbance that could potentially serve as a point source for deeply sourced or recently
286 excavated sediment, which would therefore not be indicative of long-term background surface
287 erosion rates (e.g., gravel pits, quarrying; Fig. 2). Sampled basins were small enough to be
288 occupied by one primary land use: forested, agricultural, or urban (classifications derived from a
289 national land cover database, accessed on 21 July 2011; NOAA, 2010).

290 Alluvium was collected from streambeds or point bars and sieved in the field to the 250–
291 850 μm grain-size fraction for both $^{10}\text{Be}_i$ and $^{10}\text{Be}_m$ analyses. Several studies have shown that the
292 concentration of $^{10}\text{Be}_m$ can be higher on smaller grain sizes than on coarser material (Dannhaus
293 et al., 2018; Pavich et al., 1985; Singleton et al., 2015), which could lead to biases in
294 determinations of the denudation rate (von Blanckenburg et al., 2012); however, $^{10}\text{Be}_m$ grain-size
295 dependencies are minimized when $^{10}\text{Be}_m$ is normalized to $^{9}\text{Be}_{\text{reac}}$ (Singleton et al., 2015; von
296 Blanckenburg et al., 2012; Wittmann et al., 2012). Samples from each basin were used for $^{10}\text{Be}_m$
297 analyses ($n = 70$); sufficient amounts of pure quartz were extracted from samples for the majority
298 of basins ($n = 62$), from which $^{10}\text{Be}_i$ was used to calculate basin-averaged, long-term erosion
299 rates (Bierman and Steig, 1996; Brown et al., 1995; Granger et al., 1996).

300 The ^{10}Be samples were prepared for isotope dilution chemistry using a ^9Be carrier
301 solution that was added during sample processing. Beryllium was isolated and purified using the
302 methods of Corbett et al. (2016) for $^{10}\text{Be}_i$ and Stone (1998) for $^{10}\text{Be}_m$ samples. Beryllium isotope
303 ratios ($^{10}\text{Be}/^{9}\text{Be}$) were measured by accelerator mass spectrometry (AMS) at the Lawrence
304 Livermore National Laboratory (Rood et al., 2010), blank-corrected, and normalized to the
305 07KNSTD standard assuming a standard ratio of 2.85×10^{-12} (Nishiizumi et al., 2007; Table
306 DR2). We summarized $^{10}\text{Be}_i$ production across each sampled basin to a single point in space and
307 calculated basin-scale erosion rates from $^{10}\text{Be}_i$ data using the CRONUS online calculator (Balco
308 et al., 2008; main calculator, version 2.1) using a global $^{10}\text{Be}_i$ production rate and scaling
309 schemes of Lal (1991) and Stone (2000). Typically, cosmogenic erosion rates are representative
310 of all upstream catchment areas contributing sediment to that sample, but here we present
311 erosion rates that reflect the un-overlapped portions of nested catchments following Granger et
312 al. (1996); we refer to these erosion rates as unnested erosion rates, and we use the unnested rates
313 for all following calculations and statistics (Table DR3).

314 In addition to $^{10}\text{Be}_i$ erosion rates, deriving erosion rates from single measurements of
315 $^{10}\text{Be}_m$ has previously been proposed (Brown et al., 1988), but the idea was supplanted by the
316 more straightforward $^{10}\text{Be}_i$ erosion rate technique described above. Recently, however, deriving
317 erosion rates from $^{10}\text{Be}_m$ has been revisited (von Blanckenburg et al., 2012; Willenbring and von
318 Blanckenburg et al., 2010) using the following equation:

$$319 E = \frac{^{10}\text{Be}_i F_{\text{met}}}{[^{10}\text{Be}_m]}, \quad (1)$$

320 Here, $^{10}\text{Be}_i F_{\text{met}}$ is the $^{10}\text{Be}_m$ delivery rate (atoms $\text{cm}^{-2} \text{yr}^{-1}$) and $[^{10}\text{Be}_m]$ is the concentration of
321 $^{10}\text{Be}_m$ (atoms g^{-1}). E , the erosion rate, is expressed in units of $\text{g cm}^{-2} \text{yr}^{-1}$; however, we convert

322 these units and present E in units of $\text{Mg km}^{-2} \text{ yr}^{-1}$. E is also commonly expressed in units of m
 323 m.y.^{-1} by taking the quotient of E and bulk rock density (e.g. 2.7 g cm^{-3}), then converting units.

324 Denudation rates, total mass loss from both chemical and physical processes, can also be
 325 derived using $^{10}\text{Be}_m$ by normalizing $^{10}\text{Be}_m$ measurements to $^9\text{Be}_{\text{reac}}$ measurements, using a
 326 rearrangement of von Blanckenburg et al.'s (2012) mass-balanced Equation 12:

$$327 D = \frac{^{10}\text{Be}F_{\text{met}} \times \left(\frac{[{}^9\text{Be}]_{\text{min}}}{[{}^9\text{Be}]_{\text{reac}}} + 1 \right)}{[{}^9\text{Be}]_{\text{parent}} \times \left(\frac{^{10}\text{Be}_m}{^9\text{Be}_{\text{reac}}} \right)}, \quad (2)$$

328 where D is the denudation rate ($\text{g cm}^{-2} \text{ yr}^{-1}$), and $[{}^9\text{Be}]_{\text{parent}}$, $[{}^9\text{Be}]_{\text{min}}$, and $[{}^9\text{Be}]_{\text{reac}}$ are the native,
 329 mineral, and reactive concentrations of ^9Be , respectively (atoms g^{-1}). Here, we convert units and
 330 present D in units of $\text{Mg km}^{-2} \text{ yr}^{-1}$. Importantly, Equation 2 assumes that there is minimal mass
 331 flux of ^9Be dissolved in stream water, an assumption that is likely valid considering that average
 332 pH measurements from nine of the ten sampled streams with USGS gauging stations (Fig. 2) are
 333 between 7.0–8.1 (average measurement period: 30 yrs; average number of pH measurements per
 334 station: 270; no pH data were available for the tenth gauging station, 01650500). In contrast, pH
 335 < 4 is typically required to strip all Be from grain coatings (Åström et al., 2018; Graly et al.,
 336 2010; Willenbring et al., 2010). Data for these nine USGS gauging stations were accessed
 337 through the USGS National Water Information System
 338 (<https://maps.waterdata.usgs.gov/mapper>). Because there are likely negligible amounts of
 339 $^9\text{Be}_{\text{parent}}$ being lost in the dissolved phase, $^{10}\text{Be}_m/{}^9\text{Be}_{\text{reac}}$ denudation rates can be directly
 340 compared to $^{10}\text{Be}_i$ erosion rates assuming that soil depths are similar to or less than the depth of
 341 cosmic ray penetration (Pavich, 1989, 1990). If the weathering front is deeper than a few meters,
 342 $^{10}\text{Be}_m/{}^9\text{Be}_{\text{reac}}$ denudation rates should exceed $^{10}\text{Be}_i$ erosion rates.

343 von Blanckenburg et al.'s (2012) mass-balance approach, and thus Equation 2 can only
 344 be used if certain criteria are met: (1) $^{10}\text{Be}F_{\text{met}}$ is known or can be reasonably estimated, (2)
 345 $[{}^9\text{Be}]_{\text{parent}}$ is known or can be assumed, (3) the $^{10}\text{Be}_m/{}^9\text{Be}_{\text{reac}}$ ratio has achieved equilibrium in the
 346 weathering zone, (4) the $^{10}\text{Be}_m/{}^9\text{Be}_{\text{reac}}$ ratio of beryllium adhered to sediment is in equilibrium
 347 with the $^{10}\text{Be}_m/{}^9\text{Be}_{\text{reac}}$ ratio of beryllium dissolved in a river basin, (5) the basin area is of
 348 sufficient size to average out natural variations of the $^{10}\text{Be}_m/{}^9\text{Be}_{\text{reac}}$ ratios, (6) radioactive decay
 349 of $^{10}\text{Be}_m$ is negligible, (7) $^{10}\text{Be}_m$ and $^9\text{Be}_{\text{reac}}$ can be extracted from sediment, and (8) the fractional
 350 flux of ^9Be released from bedrock parent material, von Blanckenburg et al.'s (2012) (${}^9\text{Be}f_{\text{reac}} +$
 351 ${}^9\text{Be}f_{\text{diss}}$) term, is known.

352 Information presented below suggest that each of the criteria listed above are reasonably
 353 met in the Potomac River basin. (1) The $^{10}\text{Be}_m$ flux rate we use, $^{10}\text{Be}F_{\text{met}}$ ($\text{atoms cm}^{-2} \text{ yr}^{-1}$), is
 354 derived from observed relationships among measured deposition rates, precipitation rate (P ,
 355 in cm yr^{-1}), and latitude (L) (Graly et al., 2011) such that:

$$356 {}^{10}\text{Be}F_{\text{met}} = P \times \frac{1.44}{\left(1 + e^{\left(\frac{(30.7 - L)}{4.36} \right)} \right) + 0.63}. \quad (3)$$

357 Values of $^{10}\text{Be}F_{\text{met}}$ for individual catchments range from $1.5\text{--}2.5 \times 10^6 \text{ atoms cm}^{-2} \text{ yr}^{-1}$, with an
 358 average of $2.0 \times 10^6 \text{ atoms cm}^{-2} \text{ yr}^{-1}$, which is consistent with an average Holocene $^{10}\text{Be}F_{\text{met}}$ -value
 359 of $2.0 \times 10^6 \text{ atoms g}^{-1} \text{ yr}^{-1}$ (Heikkilä and von Blanckenburg, 2015). (2) We did not directly
 360 measure $[{}^9\text{Be}]_{\text{parent}}$, and we follow von Blanckenburg et al. (2012) in assuming an average crustal
 361 $[{}^9\text{Be}]_{\text{parent}}$ concentration of 2.5 ppm. (3) The catchments that our samples come from are long-
 362 established and we thus assume that the $^{10}\text{Be}_m/{}^9\text{Be}_{\text{reac}}$ ratio has equilibrated in the weathering
 363 zone. (4) There is minimal dissolved beryllium considering regional river water pH values at
 364 stream gauging stations and thus the $^{10}\text{Be}_m/{}^9\text{Be}_{\text{reac}}$ ratios are fully equilibrated between reactive

365 and dissolved phases. (5) The measured catchments are sufficiently large (5–30,000 km²) to
 366 ensure sediment mixing. (6) Little ¹⁰Be_m has decayed between its production and sample
 367 collection because rates of erosion limit regolith residence time on the landscape to much less
 368 than the half-life of ¹⁰Be. (7) The HCl leaching technique removes primarily ¹⁰Be_m and ⁹Be_{reac}
 369 from sediment grain coatings and leaches little if any ⁹Be_{min} (Greene, 2016) as indicated by
 370 minimal Si in the leachate. (8) The (⁹Be_{reac} + ⁹Be_{diss}) term is implicit in Equation 2 (above) by the
 371 following rearrangements of von Blanckenburg et al.'s (2012) Equations 9 and 10, respectively:

$$372 \quad ({}^{9\text{Be}}f_{\text{reac}} + {}^{9\text{Be}}f_{\text{diss}}) = \frac{{}^{10\text{Be}}F_{\text{met}}}{D \times [{}^9\text{Be}]_{\text{parent}} \times \left(\frac{{}^{10\text{Be}}m}{{}^9\text{Be}_{\text{reac}}}\right)}, \quad (4)$$

373 and

$$374 \quad \frac{1}{\left(\frac{[{}^9\text{Be}_{\text{min}}]}{[{}^9\text{Be}_{\text{reac}}]} + 1\right)} = \frac{{}^{10\text{Be}}F_{\text{met}}}{D \times [{}^9\text{Be}]_{\text{parent}} \times \left(\frac{{}^{10\text{Be}}m}{{}^9\text{Be}_{\text{reac}}}\right)}. \quad (5)$$

375 The right-hand sides of Equations 4 and 5 are equal; therefore:

$$376 \quad ({}^{9\text{Be}}f_{\text{reac}} + {}^{9\text{Be}}f_{\text{diss}}) = \frac{1}{\left(\frac{[{}^9\text{Be}]_{\text{min}}}{[{}^9\text{Be}]_{\text{reac}}} + 1\right)}. \quad (6)$$

377 Importantly, Equations 4–6 (above) assume there is negligible ⁹Be leaving any catchment in the
 378 dissolved phase, which reduces the denominator in von Blanckenburg et al.'s (2012) Equation 10
 379 to unity. Note that Equation 5 (above) is also a rearrangement of von Blanckenburg et al.'s
 380 (2012) Equation 12, which we rearranged to solve for *D* in Equation 2 of this paper.

381 Erosion indices (EIs), calculated only for samples for which there is annual sediment load
 382 data (*n* = 10, Gellis et al., 2004) were quantified using the approach of Brown et al. (1988):

$$383 \quad EI = \frac{M \times [{}^{10}\text{Be}_m]}{A \times {}^{10\text{Be}}F_{\text{met}}}, \quad (7)$$

384 where *M* is the annual sediment load (g yr⁻¹), [¹⁰Be_m] is the isotopic concentration of ¹⁰Be_m
 385 (atoms g⁻¹), and *A* is river basin area (cm²). EIs are <1 when more ¹⁰Be_m is delivered to the basin
 386 by precipitation than is exported in sediment grain coatings; consequently, ¹⁰Be, and sediment by
 387 inference, is stored within the river basin. Conversely, EIs >1 indicate rates of ¹⁰Be_m export
 388 exceed rates of ¹⁰Be_m delivery to a basin, and this can be used to infer sediment export.

389 All ¹⁰Be_m, ⁹Be_{reac}, and ⁹Be_{min} samples were processed at the University of Vermont
 390 Cosmogenic Nuclide Laboratory (Table DR2; www.uvm.edu/~cosmolab). The ⁹Be_{reac} was
 391 measured on aliquots of all samples (*n* = 70) by inductively coupled plasma–optical emission
 392 spectrometry (ICP-OES) after being leached using heated, ultrasonic etching in 6 M HCl
 393 (Greene, 2016). The ⁹Be_{min} was extracted from already acid-etched bulk sediment from 57 of the
 394 70 basins by a multistep open-beaker hotplate procedure using concentrated H₂O₂, HNO₃, HF,
 395 and HClO₄ (see data repository for detailed sample processing procedures [see footnote 1]). ICP-
 396 OES measurements of Si in the 6 M HCl leachate, extracted at different time intervals during the
 397 leaching process, indicate that predominately ⁹Be_{reac} in grain coatings was removed and ⁹Be_{min} in
 398 primary silicate minerals was left intact (Greene, 2016).

399 Brown et al. (1988) compared the flux of ¹⁰Be_m entering drainage basins via precipitation
 400 with that adhered to sediment (as grain coatings) exiting the same basins in order to assess where
 401 sediment export or storage occurs throughout the Appalachian Mountains; they termed this ratio
 402 the erosion index (EI) and made their measurements in bulk sediment samples after testing for
 403 and finding little effect of grain size on ¹⁰Be_m concentration in one sample. Three of Brown et
 404 al.'s (1988) sampled basins are located within the Potomac River basin. Brown et al. (1988)
 405 found that, in general, EIs were very sensitive to land-use practices and that catchments draining
 406 the Piedmont region along the entire US east coast were exporting rather than storing sediment.

407 We recalculated Brown et al.'s (1988) EI values for sub-basins in the Potomac River basin using
 408 more than 20 yr of precipitation data (Hijmans et al., 2005) and basin-specific $^{10}\text{Be}_m$ delivery
 409 estimates (Eq. 3; Graly et al., 2011). We adjusted Brown et al.'s (1988) published $^{10}\text{Be}_m$
 410 concentrations by a factor of 0.9042 (see
 411 hess.ess.washington.edu/math/docs/al_be_v22/al_be_docs.html) because the reported
 412 concentrations were originally derived using the ICN/KNSTD AMS standard material, which
 413 was the primary standard material used at University of Pennsylvania at the time Brown made
 414 his measurements (Middleton et al., 1993; Nishiizumi et al., 2007); the correction factor is based
 415 on an updated ^{10}Be AMS standard value, which is consistent with a change in the accepted ^{10}Be
 416 half-life (Nishiizumi et al., 2007).

417 To understand how the $^{10}\text{Be}_m$ concentration of sand-sized sediment moving through the
 418 Potomac River has changed over time (150 k.y.), we measured the concentrations of $^{10}\text{Be}_m$
 419 adhered to sediment extracted from 13 samples from the well-dated Hybla Valley sediment core
 420 (Fig. 2; Litwin et al., 2013). A similar approach was used to infer histories of sediment delivery
 421 to Chesapeake and San Francisco Bays (Helz and Valette-Silver, 1992; Valette-Silver et al.,
 422 1986; van Geen et al., 1999).

423 RESULTS

424 Long-term, background $^{10}\text{Be}_i$ -based erosion rates (Tables DR2 and DR3 [see footnote 1])
 425 for 62 Potomac River basins range from 8 to 104 $\text{Mg km}^{-2} \text{yr}^{-1}$ (3 to 39 m m.y.^{-1}) and are
 426 normally distributed according to a Shapiro-Wilk goodness-of-fit test ($p = 0.23$; Fig. 2). We used
 427 a Grubb's outlier test to identify statistical outliers, which we investigated further to determine if
 428 these samples were collected from severely disturbed basins. Although both samples POT20 and
 429 POT45 were identified as outliers, we only exclude POT20 from further analyses (including all
 430 $^{10}\text{Be}_m$ -based analyses) because it drains a large portion of agricultural research land that is
 431 heavily disturbed and because there is no certainty that all sediment derived upstream of POT20
 432 originated from within the basin, as some may have been brought in from elsewhere for research
 433 purposes. The mean $^{10}\text{Be}_i$ erosion rate for the Potomac River basin is $29.6 \pm 14.1 \text{ Mg km}^{-2} \text{yr}^{-1}$
 434 ($11.0 \pm 5.2 \text{ m m.y.}^{-1}$; 1σ , $n = 61$), and median and area-weighted mean erosion rates are within
 435 the uncertainty of the mean. At these erosion rates, material resides within the uppermost $\sim 60 \text{ cm}$
 436 of Earth's surface for $\sim 55 \text{ k.y.}$, on average, integrating over much of the last glacial-interglacial
 437 cycle. However, ten contemporary sediment yield measurements (Gellis et al., 2004) are up to
 438 ~ 10 x greater than the background sediment flux calculated from $^{10}\text{Be}_i$ (Fig. 3a). Potomac River
 439 erosion rates derived from $^{10}\text{Be}_m$ using the two-factor Equation 1 are, in nearly every case,
 440 greater than $^{10}\text{Be}_i$ erosion rates (Fig. 3b). No systematic offset exists between the two erosion
 441 rate datasets, except in the Coastal Plain where $^{10}\text{Be}_m$ erosion rates are consistently ~ 20 x greater
 442 than $^{10}\text{Be}_i$ erosion rates.

443 $^{10}\text{Be}_i$ erosion rates are correlated to mean basin slope in the Potomac River basin ($R^2 = 0.115$;
 444 $p = 0.008$); this correlation, while significant, is much weaker than the correlations
 445 between $^{10}\text{Be}_i$ erosion rates and slope found in other nonglaciated basins in the Appalachian
 446 Mountains: $R^2 = 0.54$ for the Blue Ridge Escarpment (Linari et al., 2016); $R^2 = 0.42$ for the
 447 Great Smoky Mountains (Matmon et al., 2003); $R^2 = 0.58$ for the Susquehanna River basin
 448 (Reuter, 2005). As a whole population, $^{10}\text{Be}_i$ erosion rates in the Potomac Basin ($n = 61$) are
 449 correlated with mean basin elevation ($R^2 = 0.160$; $p = 0.001$) and mean annual precipitation (R^2
 450 = 0.146; $p = 0.002$; Hijmans et al., 2005), but not with basin relief ($R^2 = 0.034$; $p = 0.156$) or
 451 basin area ($R^2 = 0.022$; $p = 0.253$); $^{10}\text{Be}_i$ erosion rates exhibit a weak but significant correlation
 452 ($R^2 = 0.065$) – after rounding ($p = 0.047$) – with latitude, which is a proxy for distance from the

453 Last Glacial Maximum ice-sheet margin. All elevation data were derived from the 90 m Satellite
 454 Radar Topography Mission dataset, downloaded from earthexplorer.usgs.gov.

455 Analysis of variance of $^{10}\text{Be}_i$ erosion rates shows no distinguishable difference in the
 456 mean erosion rate of samples from different land-use categories (Fig. 4a). Similarly, analysis of
 457 variance of $^{10}\text{Be}_i$ erosion rates in the five physiographic provinces shows that there is no
 458 statistical difference in the mean $^{10}\text{Be}_i$ erosion rates from any province (Fig. 4b). We note,
 459 however that there is a wider range in erosion rates in the Appalachian Plateau and that the $^{10}\text{Be}_i$
 460 rates from the Coastal Plain may be lower than those from other provinces since the p -value for
 461 this analysis is equal to the significance threshold after rounding ($p = 0.048$).

462 $^{10}\text{Be}_m$ concentrations and $^{10}\text{Be}_i$ concentrations are not correlated when considered as a
 463 whole population ($R^2 = 0.044, p = 0.104$; Fig. 3c). However, positive correlations between $^{10}\text{Be}_m$
 464 and $^{10}\text{Be}_i$ concentrations are observed in the Appalachian Plateau ($R^2 = 0.664, p = 0.048$), Valley
 465 and Ridge ($R^2 = 0.277, p = 0.025$), and Coastal Plain ($R^2 = 0.981, p < 0.001$) physiographic
 466 provinces (Fig. 3c). $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ are not related within statistical significance to $^{10}\text{Be}_i$ erosion
 467 rates throughout the Potomac River basin (e.g., $p > 0.05$, Fig. 5). When considering all Potomac
 468 samples, denudation rates derived from $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ ratios using Equation 2 are weakly
 469 correlated to $^{10}\text{Be}_i$ erosion rates ($R^2 = 0.208, p < 0.001$; Fig. 3d; Table DR3 [see footnote 1]); a
 470 Students t -Test shows the means of these two datasets to be similar as well ($p = 0.003$). Although
 471 weakly correlated, $^{10}\text{Be}_i$ erosion rates for individual basins are not reproduced well by
 472 $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates. We find that $^{10}\text{Be}_m$ erosion rates are often much greater than
 473 $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates.

474 EIs for samples we collected and those from three sites of Brown et al. (1988) within the
 475 Potomac River watershed are representative of the total upstream $^{10}\text{Be}_m$ isotopic balance (Brown
 476 et al., 1988). EIs calculated for the same basins sampled ~20 yr apart by us and those reported in
 477 Brown et al. (1988) are nearly equivalent (e.g., EI = 0.55 for POT06 and EI = 0.58 for USGS
 478 gauging station 01638500) and thus are reproducible over time. EIs for samples POT01–POT13
 479 range from 0.07 to 1.24, and the EIs of POT02, POT10, POT12 are within 10% of equilibrium
 480 with respect to $^{10}\text{Be}_m$ delivery and export (Fig. 6; Table DR3 [see footnote 1]). EIs for sample
 481 sites that drain large portions of the Valley and Ridge (e.g. POT01, POT06 and gauging station
 482 01638500) or small portions of the Piedmont provinces (e.g. POT 04, POT05, POT09, POT13)
 483 are generally <1 ; basins draining large portions of the Piedmont (e.g. gauging station 01643000),
 484 small basins in the Valley and Ridge (e.g. gauging station 01610200), and the Appalachian
 485 Plateau (e.g. POT11) are all >1 .

486 DISCUSSION

487 Our measurements of $^{10}\text{Be}_i$, $^{10}\text{Be}_m$, $^{9}\text{Be}_{\text{min}}$, and $^{9}\text{Be}_{\text{reac}}$ from the Potomac River basin
 488 allow us to determine the pace of landscape change ($^{10}\text{Be}_i$ erosion rates) and to revisit and
 489 evaluate three uses of $^{10}\text{Be}_m$ proposed by others (cf. Brown et al., 1988; von Blanckenburg et al.,
 490 2012; Willenbring and von Blanckenburg, 2010): as a metric of erosion ($^{10}\text{Be}_m$), as a metric of
 491 denudation ($^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$), and as a measure of landscape stability using the erosion index
 492 (sediment yield and $^{10}\text{Be}_m$). Our comparison between $^{10}\text{Be}_i$ and $^{10}\text{Be}_m$ -derived erosion and
 493 denudation metrics is an important experiment to conduct because it independently tests and
 494 applies to a new geographic region the approach suggested by von Blanckenburg et al. (2012)
 495 and implemented thus far by Dannhaus et al. (2018), Rahaman et al. (2017), and Wittmann et al.
 496 (2015).

497 Comparison of $^{10}\text{Be}_i$, $^{10}\text{Be}_m$, and $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ -inferred rates of landscape change

498 Interpreting $^{10}\text{Be}_m$ alone in measured in detrital, fluvial sediment as erosion rates using

499 eq. 1 does not produce accurate erosion rate data; in most cases, $^{10}\text{Be}_m$ overestimates, sometimes
 500 grossly, erosion rates determined using $^{10}\text{Be}_i$. Although concentrations of $^{10}\text{Be}_m$ and $^{10}\text{Be}_i$ are
 501 correlated in Potomac River sediment collected from sub-basins in the Appalachian Plateau ($n =$
 502 5), Valley and Ridge ($n = 18$), and Coastal Plain ($n = 7$), no such correlation exists in the
 503 Piedmont and the Blue Ridge provinces, nor over the Potomac Basin sample set as a whole.
 504 These data thus suggest that concentrations of $^{10}\text{Be}_m$ as we collected it (e.g. sand-only sized
 505 fraction) do not reliably represent regolith residence times. This conclusion is consistent with
 506 von Blanckenburg et al.'s (2012) advocacy of a more complex analytical approach involving the
 507 measurement of $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ and the incorporation of other assumed and measured parameters
 508 (eq. 2 and 6).

509 In contrast to the $^{10}\text{Be}_m$ system, average rates of landscape change measured using $^{10}\text{Be}_m$
 510 $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ and $^{10}\text{Be}_i$ are more similar indicating that normalization by $^{9}\text{Be}_{\text{reac}}$ is important and
 511 reasonable. However, differences between the $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ and $^{10}\text{Be}_i$ systems remain and are
 512 are most apparent in data from the Piedmont province, which differs from the other
 513 physiographic provinces in the Potomac River basin because it has very thick regolith and thus
 514 great depth to the weathering front. Measurements of $^{10}\text{Be}_i$ will underestimate the total mass lost
 515 in such landscapes because of mass loss by solution and groundwater export at depths below
 516 most *in situ* nuclide production (e.g. Riebe et al., 2001). A maximum value for solution loss
 517 below the depth of cosmic ray penetration can be approximated by the dissolved load of streams
 518 at base flow; Pavich (1990) made such calculations for the Piedmont and suggests that 10.8 Mg
 519 $\text{km}^{-2} \text{yr}^{-1}$ is lost in solution ($\sim 4 \text{ m m.y.}^{-1}$, assuming a density of 2,700 kg m^{-3}). If all solutes in the
 520 Piedmont originate below the cosmic ray penetration depth, the discrepancy between $^{10}\text{Be}_i$
 521 erosion rates and $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates in the Piedmont (30.8 $\text{Mg km}^{-2} \text{yr}^{-1}$ and 46.7 Mg
 522 $\text{km}^{-2} \text{yr}^{-1}$, respectively) can be, for the most part, resolved by adding the mass lost to solution
 523 (Pavich, 1990) to the $^{10}\text{Be}_i$ inferred erosion rate. This yields a total mass loss rate in the Piedmont
 524 (41.6 $\text{Mg km}^{-2} \text{yr}^{-1}$, dissolved load plus $^{10}\text{Be}_i$ inferred erosion rate), closer to the measured
 525 $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rate (46.7 $\text{Mg km}^{-2} \text{yr}^{-1}$).

526 The lack of consistent agreement, for almost all sub-basins, between the $^{10}\text{Be}_i$ and $^{10}\text{Be}_m$
 527 erosion rates (Fig. 3b) and between $^{10}\text{Be}_i$ erosion rates and $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates (Fig.
 528 3d) suggests that $^{10}\text{Be}_m$ methods are not accurately measuring long term rates of landscape
 529 change at the sub-basin scale, at least in the Potomac River basin. Our observation that
 530 $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates do not replicate $^{10}\text{Be}_i$ erosion rates in the Blue Ridge – the steepest
 531 of all sampled provinces, and thus the one with the thinnest soils – and that $^{10}\text{Be}_m$ erosion rates
 532 are generally much greater than $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates throughout a landscape where
 533 soils are generally thin, suggests noise in the $^{10}\text{Be}_m$ -based data sets exceeds that of the $^{10}\text{Be}_i$ -
 534 based data sets.

535 Such noise is not surprising considering the complexity of $^{10}\text{Be}_m$ behavior in the near-
 536 surface weathering system where physical, chemical, biologic, and pedogenic processes control
 537 $^{10}\text{Be}_m$ distribution over time, depth and space (Graly et al., 2010). This stands in contrast to $^{10}\text{Be}_i$,
 538 for which the production function with depth and over space and time is well constrained by the
 539 relevant nuclear physics. Below, we speculate on what factors might lead to both scatter and bias
 540 in rates derived from $^{10}\text{Be}_m$ and $^{9}\text{Be}_{\text{reac}}$ data in contrast to $^{10}\text{Be}_i$.

541

542 **Grain Size Bias**

543 Measuring $^{10}\text{Be}_m$ and $^{9}\text{Be}_{\text{reac}}$ on sand-size sediment underestimates total $^{10}\text{Be}_m$ and $^{9}\text{Be}_{\text{reac}}$
 544 because there is relationship between grain coating volume, surface area, and grain size (Graly et

545 al., 2010; Wittmann et al., 2012). In theory (von Blanckenburg et al., 2012), the ratio between the
 546 two isotopes of Be should be constant and thus denudation rates inferred from $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$
 547 should be unaffected by grain size. This assertion has been independently investigated (Singleton
 548 et al., 2016) who found that while the normalizing $^{10}\text{Be}_m$ to $^{9}\text{Be}_{\text{reac}}$ does not completely correct
 549 for grain-size biases, the bias was greatly diminished (i.e. a ~1.7 to 6-fold difference between
 550 $^{10}\text{Be}_m$ on clays compared to medium sand when uncorrected, versus a ~1.2 to 3-fold difference
 551 between $^{10}\text{Be}_m$ on clays compared to medium sand). The issue of grain size is a difficult one.
 552 Although $^{10}\text{Be}_m$ and $^{9}\text{Be}_{\text{reac}}$ can be measured in different size fractions, such data are of limited
 553 utility unless one knows the grain-size specific mass flux from a basin. Data that as far as we
 554 know are unavailable except in unique circumstances such as where all sediment leaving a basin
 555 is trapped and analyzed for grain size abundance.

556 Although grain size is doubtless important for interpreting measured $^{10}\text{Be}_m$
 557 concentrations, data suggest its effect in the Potomac basin is minimal. First, most sediment
 558 transport occurs in the Potomac region during large storms at which time sand is the most
 559 common grain-size transported (Brown et al., 1988). Consistent with other studies using the
 560 $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ technique (e.g. Dannhaus et al., 2018; Rahaman et al., 2017; Wittmann et al., 2015)
 561 we sampled the most commonly transported grain size, in this case, sand. Second, Brown et al.
 562 (1988) only found 1% of total $^{10}\text{Be}_m$ was exported in the $<250\text{ }\mu\text{m}$ grain-size fraction of an
 563 Appalachian stream similar to those we sampled. Third, in this study we normalized $^{10}\text{Be}_m$ using
 564 ^{9}Be measurements, which should have significantly reduced grain size biases (von Blanckenburg
 565 et al., 2012).

566 **Sample Processing Bias**

567 In extracting $^{10}\text{Be}_m$ and $^{9}\text{Be}_{\text{reac}}$ from sediment grain coatings, we pulverized the sediment
 568 and performed a 6 M HCl leach on each sample. Using a strong acid leach on pulverized sample
 569 material differs from other studies that leach original material using both 0.5 M HCl and
 570 hydroxylamine-hydrochloride solutions (Wittmann et al., 2012). Laboratory measurements of the
 571 solute taken from our samples at intervals during the etching process demonstrate that 6 M HCl
 572 did not remove any significant amount of Si. Silica in solution would be an indication that
 573 silicate minerals, including quartz, were dissolved to some degree, releasing $^{9}\text{Be}_{\text{min}}$ into solution;
 574 thus, we conclude little if any $^{9}\text{Be}_{\text{min}}$ was incorporated into the $^{9}\text{Be}_{\text{reac}}$ measurements (Greene,
 575 2016; Singleton et al. 2016).

576 **Unconstrained $^{9}\text{Be}_{\text{parent}}$ or $^{10}\text{Be}_m F_{\text{met}}$**

577 We did not measure the amount of naturally-occurring $^{9}\text{Be}_{\text{parent}}$ in bedrock samples from
 578 all lithologies throughout the Potomac River basin and no such data are available. Rather, we
 579 follow previous studies in assuming a bulk crustal $^{9}\text{Be}_{\text{parent}}$ concentration (von Blanckenburg et
 580 al., 2012). Doing so introduces uncertainty into the value of the denominator in Equation 2.

581 It is possible that the $^{10}\text{Be}_m$ delivery rate ($^{10}\text{Be}_m F_{\text{met}}$) has changed through time. However,
 582 average Holocene $^{10}\text{Be}_m F_{\text{met}}$ rates (Heikkila and von Blanckenburg, 2015) are similar to the $^{10}\text{Be}_m F_{\text{met}}$
 583 rates we calculated for our field area following Graly et al. (2011) which is based on
 584 contemporary $^{10}\text{Be}_m$ flux data. If our data sets are affected by inaccurate representations of
 585 $^{9}\text{Be}_{\text{parent}}$ or $^{10}\text{Be}_m F_{\text{met}}$, all samples would be affected, and the scatter in our data sets would be
 586 unchanged.

587 **Effects of colonial-era land use in the Potomac Region**

588 European-American colonial-era land use is the most likely and reasonable explanation
 589 for the disagreement between $^{10}\text{Be}_i$ and $^{10}\text{Be}_m$ erosion rates, between $^{10}\text{Be}_m$ erosion rates and
 590 $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates, and between $^{10}\text{Be}_i$ sediment fluxes, $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates,

591 and historical sediment yields.

592 Valette-Silver et al. (1986) showed that topsoil erosion associated with European-
 593 American deforestation (Brush, 2009) led to deposition of post-colonial sediment in the northern
 594 arm of Chesapeake Bay, fed by the Susquehanna River. $^{10}\text{Be}_m$ concentrations associated with
 595 these post-colonial sediments ($7\text{--}11 \times 10^8$ atoms g^{-1}) were well above pre-colonial $^{10}\text{Be}_m$
 596 concentrations (2×10^8 atoms g^{-1}). After colonial-era erosion removed many centimeters of near-
 597 surface topsoil (Costa, 1975), likely containing the highest $^{10}\text{Be}_m$ concentrations (Graly et al.,
 598 2010), concentrations of $^{10}\text{Be}_m$ on sediment delivered to Chesapeake Bay dropped to $\sim 5 \times 10^8$
 599 atoms g^{-1} . Few $^{10}\text{Be}_m$ measurements in our Potomac River samples have concentrations $<2 \times 10^8$
 600 atoms g^{-1} or $>5 \times 10^8$ atoms g^{-1} .

601 Considering that similar land-use practices were used in both the Susquehanna and
 602 Potomac watersheds during the colonial era, it is likely that that ^{10}Be -enriched topsoil in the
 603 basins delivering sediment to our sample sites was eroded just after settlement. $^{10}\text{Be}_m$
 604 concentration in river sediment now reflects $^{10}\text{Be}_m$ -depleted parts of soil profiles, the highest
 605 concentration material having been swept downstream and into the Bay during and after colonial
 606 and post-colonial land clearance.

607 A sediment core collected at the mouth of Chesapeake Bay, downstream from the tidal
 608 Potomac River, however, shows $^{10}\text{Be}_m$ measurements of $<5 \times 10^8$ atoms g^{-1} (Helz and Valette-
 609 Silver, 1992) – i.e. no colonial-era $^{10}\text{Be}_m$ spike is observed. Thus, if $^{10}\text{Be}_m$ in topsoil was eroded
 610 from sampled basins, much of the $^{10}\text{Be}_m$ has not yet reached the mouth of Chesapeake Bay, >200
 611 km downstream from the mouth of the non-tidal Potomac River. We suggest that the lost $^{10}\text{Be}_m$
 612 is likely stored within river sub-basins, both behind milldams in the field area (Merritts et al.,
 613 2011; Walter and Merritts, 2008) and on colluvial footslopes (Reusser et al., 2015), an
 614 interpretation that is strongly supported by erosion indices derived for much of our field area that
 615 indicate sediment storage (Fig. 6). Eroded topsoil sediment and the $^{10}\text{Be}_m$ adhered to it may
 616 reside near the mouth of the Potomac River, much like the $^{10}\text{Be}_m$ spike Valette-Silver et al.
 617 (1986) identified for Chesapeake Bay was found proximal to the mouth of the Susquehanna
 618 River; however, this hypothesis remains untested.

619 The lack of clear and expected anticorrelation between the $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ ratios and $^{10}\text{Be}_i$
 620 erosion rate data sets in the Potomac River basin (Fig. 5), the dissimilarity between $^{10}\text{Be}_m$ and
 621 $^{10}\text{Be}_i$ erosion rates, and between $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates and $^{10}\text{Be}_i$ erosion rates may
 622 reflect noise inherent to the $^{10}\text{Be}_m$ system, the limited dynamic range of erosion rates (meters to
 623 tens of meters per million years) in the Potomac basin, and the lack of grainsize-specific isotopic
 624 and sediment transport flux data. A similar comparative analysis in the much larger Amazon
 625 Basin, where erosion rates vary over three orders of magnitude, showed what appeared to be a
 626 more pronounced inverse relationship between $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ ratios and $^{10}\text{Be}_i$ physical sediment
 627 fluxes as well as other metrics of erosion (Fig. 6 in von Blanckenburg et al., 2012). However, the
 628 scatter of $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ sediment generation rates in the Potomac River basin (Fig. 3d) is similar
 629 to scatter of $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ sediment generation rates within the same range ($10\text{--}100 \text{ Mg km}^{-2} \text{ yr}^{-1}$)
 630 for various river basins in North America (Fig. 3f). Together, these findings suggest that the
 631 scatter within the $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ data set is inherent to the interpretation of $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ ratios as
 632 denudation rates. This is a key finding shared between our work and previous studies and
 633 suggests that the meaningful application of $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates will be limited to field
 634 areas where denudation varies by orders of magnitude. In that case, the relative magnitude of
 635 landscape change between one part of a landscape and another can be detected, even if
 636 $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ rates from specific catchments do not consistently replicate $^{10}\text{Be}_i$ rates.

637 The overall agreement between mean $^{10}\text{Be}_i$ erosion rates and $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation
 638 rates, when considering the Potomac River basin sample set as a whole, suggests that $^{10}\text{Be}_m$
 639 when normalized to $^{9}\text{Be}_{\text{reac}}$ could be useful at a broad scale for understanding the pattern and
 640 tempo of landscape change in quartz-poor regions where $^{10}\text{Be}_i$ is not suitable (e.g. Dannhaus et
 641 al., 2018). However, until $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates can be shown to routinely replicate
 642 $^{10}\text{Be}_i$ -determined rates in sub-basins where both techniques should be expected to track the same
 643 landscape dynamics (i.e. landscapes with soil depths equal to or shallower than the spallogenic
 644 cosmogenic nuclide production depth), $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates at the sub-basin scale
 645 appear to have limited utility. Based on the Potomac River data set, it appears that applying the
 646 $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rate technique to landscapes with intensive human land-use histories
 647 (most of the currently developed world where land clearance and intensive agriculture have been
 648 practiced) is problematic.

649 Long-Term, Background Erosion Rates and Landscape Change

650 Using the data presented here, sediment dynamics in the Potomac River watershed and
 651 sediment delivery to Chesapeake Bay, can now be assessed in the context of long-term landscape
 652 change rates within the river catchment. Measurements of $^{10}\text{Be}_i$ show that the Potomac River
 653 basin erodes slowly over millennial timescales ($11.0 \pm 5.2 \text{ m m.y.}^{-1}$, assuming a rock density of
 654 $2,700 \text{ kg m}^{-3}$), closely replicating the range of basin-averaged erosion rates from the nearby
 655 Shenandoah National Park ($12.2 \pm 4.6 \text{ m m.y.}^{-1}$; Duxbury et al., 2015). However, the Potomac
 656 River basin is, on average, eroding at nearly half the pace as unglaciated parts of the
 657 Susquehanna River basin to the north (19.7 m m.y.^{-1} ; Reuter, 2005), which is interesting because
 658 the Susquehanna basin is underlain by the same physiographic provinces and bedrock
 659 lithologies, and has experienced a similar history of human land use. The two-fold difference in
 660 average long-term erosion rates between the Potomac and Susquehanna River watersheds may
 661 reflect either a base-level change affecting only the Susquehanna (i.e. Miller et al., 2013) or the
 662 closer proximity of many of the Susquehanna sub-basins to the former Laurentide glacial margin,
 663 where they experienced more intense periglacial weathering (Clark and Ciolkosz, 1988; Denn et
 664 al., 2018; Marshall et al., 2015; Whittecar and Ryter, 1992). We consider the latter of these
 665 explanations to be dominant as we interpret the positive relationship between erosion rates and
 666 latitude in the Potomac basin (Fig. 2) to reflect increased periglacial activity closer to the former
 667 glacial margin.

668 $^{10}\text{Be}_i$ -determined erosion rates are similar to but somewhat lower than rock exhumation
 669 rates measured using apatite and zircon fission-track and (U-Th)/He dating techniques (Naeber et
 670 al., 2016; Reed et al., 2005; $\sim 15\text{--}25 \text{ m m.y.}^{-1}$) over $>10^6 \text{ yr}$ timescales in the Potomac region of
 671 the central Appalachian Mountains. From this comparison of erosion rates, we infer that the pace
 672 of landscape denudation in the Potomac River basin is similar when integrated over $10^4\text{--}10^6 \text{ yr}$
 673 timescales. The observed consistency of erosion and denudation rates over different timescales is
 674 similar to that found by Matmon et al. (2003) in the Great Smoky Mountains further to the south,
 675 suggesting long-term stable rates of landscape change over time throughout the non-glaciated
 676 Appalachian Mountains.

677 A comparison of $^{10}\text{Be}_i$ basin-averaged and outcrop-specific erosion rates suggests that
 678 relief is generally not being generated throughout most of the Potomac River basin, supporting
 679 our interpretation of long-term stable rates of landscape change. Along with data from this study,
 680 $^{10}\text{Be}_i$ erosion rates from basins draining the western flank of the Blue Ridge in Shenandoah
 681 National Park (Duxbury et al., 2015) and from bedrock outcrops along ridgelines throughout the
 682 Potomac River watershed (Duxbury et al., 2015; Hancock et al., 2015; Portenga et al., 2013)

683 show that average erosion rates of basins and outcrops in the Valley and Ridge and the Blue
 684 Ridge Provinces are similar within 1σ uncertainties (Fig. 7). Outcrop erosion rates from the
 685 Appalachian Plateau are not available for the Potomac River watershed; however, $^{10}\text{Be}_i$ erosion
 686 rates of outcrops west of the continental divide in Dolly Sods, West Virginia (Hancock and
 687 Kirwan, 2007), are similar to outcrop erosion rates to the north in the Appalachian Plateau
 688 Province within the Susquehanna River basin (Portenga et al., 2013; Reuter, 2005), and so we
 689 consider these reasonable estimates of rock erosion in the Appalachian Plateau of the Potomac
 690 River watershed. When bedrock erosion rates from Dolly Sods and the Susquehanna
 691 Appalachian Plateau are assumed representative of outcrop erosion in the Potomac Appalachian
 692 Plateau, we identify and interpret the largest difference between outcrop and basin-averaged
 693 $^{10}\text{Be}_i$ erosion rates in the Potomac River watershed to reflect relief generation in the Appalachian
 694 Plateau (Fig. 7). Our observations of erosion rate difference and inferred relief generation in the
 695 Potomac Appalachian Plateau are consistent with base-level fall following stream capture west
 696 of the Blue Ridge and knickpoint retreat into headwater basins along the continental divide, as
 697 has been demonstrated for the central Appalachian Mountains (Miller et al., 2013; Naeser et al.,
 698 2016; Prince et al., 2010, 2011).

699 Erosion Indices

700 EIs of distal headwater basins indicate that more $^{10}\text{Be}_m$ is being exported from the
 701 Appalachian Plateau Province than is being retained on the landscape. This observation supports
 702 findings presented in this study that relief is being produced in the Appalachian Plateau Province
 703 and is consistent with Miller et al.'s (2013) findings that stream incision in the central
 704 Appalachian Mountains is greatest in headwater catchments along the continental divide.
 705 Conversely, EIs of the large basins (samples POT01 and POT06/USGS 01638500) indicate that
 706 $^{10}\text{Be}_m$ is being retained within the Potomac River basin. Since there are few large dams on the
 707 main branch of the Potomac River to trap sediment (Gerhart, 1991), we suggest this $^{10}\text{Be}_m$ is
 708 likely stored in the floodplain or in colluvium below hillslopes; alternatively, $^{10}\text{Be}_m$ -bearing
 709 sediment eroded from headwater streams may have become trapped behind numerous small
 710 milldams as legacy sediment (Merritts et al., 2011; Walter and Merritts, 2008) and is now being
 711 released, or will be released in the future, to the larger Potomac River basin as these milldams
 712 deteriorate. Basins with high EIs and high long-term erosion rates (e.g., POT11, USGS gauging
 713 station 01610200) are those where increased sediment delivery reflects downcutting by
 714 headwaters of the Potomac River watershed. High EIs in basins corresponding to low long-term
 715 sediment yields (POT13) reflect agricultural land use where topsoil, having long near-surface
 716 residence time (i.e., low sediment yields derived from high $^{10}\text{Be}_i$ concentrations), is eroded and
 717 carries with it large amounts of $^{10}\text{Be}_m$. $^{10}\text{Be}_m$ retention ($EI < 1$) seems also to occur in urban
 718 centers around Washington, DC (POT02–POT05) – a perplexing finding considering that
 719 sediment yields from local urban catchments are some of the highest in the Chesapeake Bay
 720 watershed (Gellis et al., 2017). The possibility of significant amounts of foreign sediment being
 721 brought into, or native sediment being exported from urban sampled basins cannot be excluded,
 722 though it is difficult to quantify such activities.

723 The equivalence between the EIs calculated for the main branch of the Potomac River
 724 from this study and from Brown et al. (1988; $EI = 0.55$ for POT06 and $EI = 0.58$ for USGS
 725 gauging station 01638500, respectively) is notable, considering that we only used the 250–850
 726 μm grain size whereas Brown et al. (1988) used unsieved samples. Such a similarity between EIs
 727 supports our earlier assertion that our $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ data set is not significantly affected by grain-
 728 size bias and shows replication in EIs from samples collected decades apart.

Because paleo-sediment loads and $^{10}\text{Be}_m$ paleo-flux rates are not known for the 13 samples collected from the Hybla Valley sediment core, past EIs for the whole of the Potomac River cannot be derived. However, we can still infer past landscape dynamics by assessing how measured $^{10}\text{Be}_m$ concentrations change through time in the Hybla Valley core (Fig. 8; cf. Helz and Valette-Silver, 1992; Valette-Silver et al., 1986; van Geen et al., 1999). If we assume $^{10}\text{Be}_m$ flux rates during the late Pleistocene have fluctuated around an average value, $^{10}\text{Be}_m$ concentrations on sediment delivered to the lower Potomac River would only increase significantly as a result of increased topsoil erosion rates, supplying more sediment to Chesapeake Bay (i.e. Valette-Silver et al., 1986). Under this assumption, the long-term trend of $^{10}\text{Be}_m$ concentrations from the Hybla Valley suggest that soil erosion was more intense and sediment delivery to the Bay increased during cold glacial periods over the last \sim 100 k.y. as indicated by decreases in abundances of oak (*Quercus* spp.), which thrives in relatively warmer climates (Litwin et al., 2013). Relative to modern $^{10}\text{Be}_m$ concentrations, low $^{10}\text{Be}_m$ concentrations are observed at ca. 80–70 ka, when oak abundances were at their greatest, indicating the warmest climate. Similarly, low $^{10}\text{Be}_m$ concentrations are observed at the top of the Hybla Core, perhaps reflecting the current interglacial warming trend. While the inferred increases in $^{10}\text{Be}_m$ and sediment export during colder climates was likely facilitated by increased periglacial activity, changes in $^{10}\text{Be}_m$ concentrations over the last \sim 100 k.y. may reflect changes in storm frequency and/or magnitude over glacial/interglacial cycles. Modern $^{10}\text{Be}_m$ concentrations from the most downstream Potomac River sample (POT01) are similar in magnitude to van Geen et al.'s (1999) observations at the mouth of Chesapeake Bay, and may reflect intensive colonial-era land use, depletion of topsoils, and exhaustion of upstream $^{10}\text{Be}_m$ soil inventories, as discussed above.

Implications of the Isotopic Data for Contextualizing Sediment Delivery to Chesapeake Bay

Efforts to restore the ecology of Chesapeake Bay to a pre-disturbance condition highlight the importance of reducing sediment delivery to the Bay (Hassett et al., 2005; Langland and Cronin, 2003), but to what level? Total maximum daily loads (TMDLs) are regulatory benchmarks for sediment delivery, derived from a combination of measured sediment yields in less disturbed basins and various land-use models (U.S. EPA, 2010). Currently, the target suspended sediment TMDL for the whole of the Potomac River basin is $24.3 \text{ Mg km}^{-2} \text{ yr}^{-1}$ (U.S. EPA, 2010), which is within the uncertainties of the long-term $^{10}\text{Be}_i$ -derived sediment flux ($29.6 \pm 14.1 \text{ Mg km}^{-2} \text{ yr}^{-1}$, 1σ). Because the $^{10}\text{Be}_i$ -inferred sediment flux for the Potomac River integrates over tens of thousands of years, it is more representative of background sediment delivery from the Potomac River to Chesapeake Bay than shorter-term sediment yields (e.g., Gellis et al., 2004) or sediment deposition rates calculated from sediment cores extracted from the Bay (e.g., Brush, 1984; Cooper and Brush, 1991, 1993; Cronin et al., 1999, 2000; Valette-Silver et al., 1986).

CONCLUSIONS

The $^{10}\text{Be}_i$ -derived erosion and thus sediment flux rates in this study provide limits on the long-term delivery rate of mass from the Potomac River to Chesapeake Bay. We find that average $^{10}\text{Be}_i$ erosion rates and $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ -derived denudation rates are similar. However, $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates do not replicate $^{10}\text{Be}_i$ -based erosion rates well when comparing sub-basins; similarly, we find that on average $^{10}\text{Be}_m$ erosion rates are greater than $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates, which is not possible and thus likely reflects grainsize bias in non-normalized $^{10}\text{Be}_m$ concentrations. Although the $^{10}\text{Be}_m$ erosion rate and $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rate technique may be useful for understanding overall rates of landscape change in quartz-poor

775 regions, the techniques does not replicate $^{10}\text{Be}_i$ -based data well in the Potomac River basin. It
 776 appears that land use impacts of the past centuries removed sediment with the highest $^{10}\text{Be}_m$
 777 concentrations biasing the calculations. This is likely to pose a complication with $^{10}\text{Be}_m$ data
 778 worldwide.

779 New $^{10}\text{Be}_i$ erosion rates for the Potomac River in this study are consistent with those
 780 presented in other $^{10}\text{Be}_i$ studies throughout the central Appalachian Mountains. Differences
 781 between basin-averaged and outcrop erosion are limited to headwaters in the Appalachian
 782 Plateau region, where erosion indices indicate net sediment export and where knickpoints are
 783 propagating into distal headwater basins. Quaternary sediment delivery from the Potomac River
 784 basin, represented by $^{10}\text{Be}_m$ concentrations in the Hybla Valley core, appears to reflect regional
 785 climate change such that more erosion of $^{10}\text{Be}_m$ -rich topsoils occurs during colder climatic
 786 periods, at least over the last 150 k.y. Over modern timescales, we find that sediment yields still
 787 greatly exceed background rates of sediment flux from the Potomac River to Chesapeake Bay,
 788 and until these rates are reduced, the ecological health of the Bay will not recover to a pre-
 789 disturbance condition. TMDLs regulating sediment delivery from the Potomac River to
 790 Chesapeake Bay are set to an appropriate level if the overall goal is to reduce sediment delivery
 791 to background rates.

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1172 Figure 1. The location of the Potomac River basin (gray), situated within the Chesapeake Bay
1173 drainage basin (bold black line) on the east coast of the United States. Extent of the Last Glacial
1174 Maximum (LGM) Laurentide ice sheet is indicated by the stippled gray line. Base map is a world
1175 shaded relief map, produced and provided open-access by ESRI. State abbreviations: NY—New
1176 York, PA—Pennsylvania, WV—West Virginia, VA—Virginia, DC—District of Columbia,
1177 MD—Maryland, DE—Delaware, NJ—New Jersey.

1178 Figure 2. Geographical distribution of $^{10}\text{Be}_i$ erosion rates within the Potomac River basin and its
1179 contributing tributaries. Open circles are sample sites with $^{10}\text{Be}_m$ data, but not $^{10}\text{Be}_i$, $^{9}\text{Be}_{\text{reac}}$, or
1180 $^{9}\text{Be}_{\text{min}}$ data; circles with bull's-eye are sites where $^{10}\text{Be}_i$, $^{10}\text{Be}_m$, $^{9}\text{Be}_{\text{reac}}$, and $^{9}\text{Be}_{\text{min}}$ were
1181 measured. Numbers next to sample sites designate the sample ID following the format POTxx.
1182 White star is the location of the Hybla Valley sediment core. Black squares are cities: B—
1183 Baltimore, Maryland (MD); C—Cumberland, MD; H—Harrisonburg, Virginia (VA); HF—
1184 Harpers Ferry, West Virginia (WV); W—Washington, DC. State abbreviations: PA—
1185 Pennsylvania, WV—West Virginia, VA—Virginia, MD—Maryland. Dashed lines represent the
1186 boundaries between physiographic provinces
1187 (water.usgs.gov/GIS/metadata/usgswrd/XML/physio.xml), and the dotted lines are state
1188 boundaries. Inset figure shows distribution of sediment generation rates derived from $^{10}\text{Be}_i$

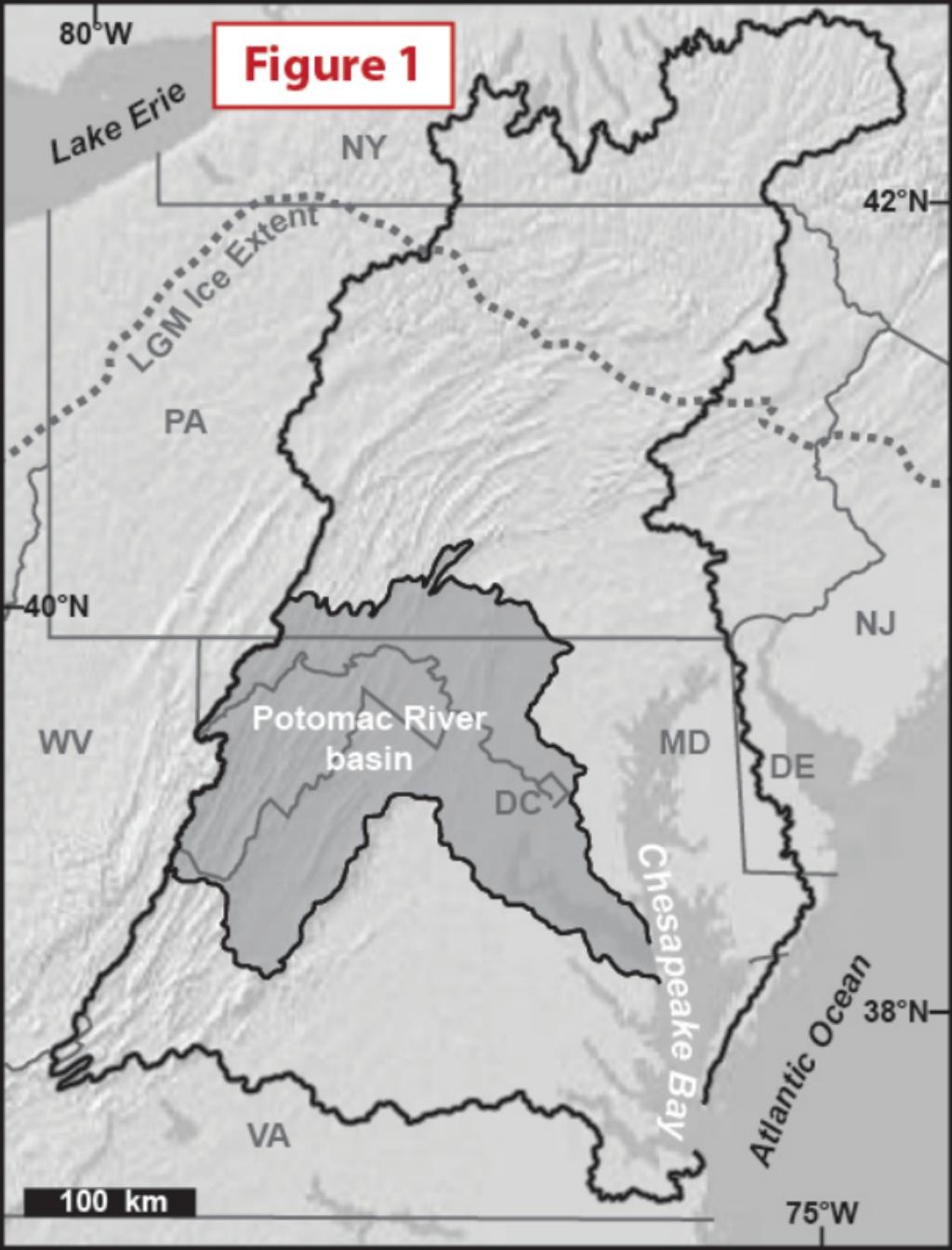
1189 (black circles), including two statistical outliers (x), which were investigated for the possible
 1190 introduction of deep-seated sediment introduced to the sample by landscape disturbance; only
 1191 POT20 (gray outlier) showed signs of heavy landscape disturbance, and thus it is not included in
 1192 any analyses. Box plot box is bounded by the 25th and 75th quartiles, and the vertical line in the
 1193 box is the median; box plot whiskers encompass all data between the 25th quartile minus $1.5 \times$
 1194 the interquartile range and the quartiles plus $1.5 \times$ the interquartile range. Base map is a world
 1195 shaded relief map, produced and provided open-access by ESRI.

1196 Figure 3. (A) Plot of historical sediment yield measured at U.S. Geological Survey gauging
 1197 stations (Gellis et al., 2004) compared to $^{10}\text{Be}_i$ -derived sediment fluxes from river sediment
 1198 collected at the same gauging stations. Contemporary sediment yields are up to $10 \times$ greater than
 1199 $^{10}\text{Be}_i$ sediment fluxes. (B) Comparison of $^{10}\text{Be}_m$ versus $^{10}\text{Be}_i$ erosion rates shows that $^{10}\text{Be}_m$
 1200 erosion rates do not replicate well the better-understood $^{10}\text{Be}_i$ erosion rates and that $^{10}\text{Be}_m$
 1201 erosion rates are nearly all unsystematically greater than $^{10}\text{Be}_i$ erosion rates, except in the Coastal
 1202 Plain. (C) Concentrations of $^{10}\text{Be}_m$ and $^{10}\text{Be}_i$ are positively and significantly correlated in the
 1203 Appalachian Plateau, the Valley and Ridge, and in the Coastal Plain physiographic provinces,
 1204 but not through the overall data set ($R^2 = 0.044$, $p = 0.104$). (D) Plot of $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation
 1205 rates versus $^{10}\text{Be}_i$ -derived sediment fluxes for the Potomac River. The two data sets are weakly
 1206 correlated (curved dashed black line in log-log space, but denudation rates from individual basins
 1207 do not replicate $^{10}\text{Be}_i$ -based sediment fluxes well. (E) Sediment fluxes derived from $^{10}\text{Be}_m$ -only
 1208 (eq. 1) versus $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ denudation rates (eq. 2). Denudation rates and sediment fluxes are
 1209 unable to replicate each other, basin-by-basin, even though in a landscape where soils are thin
 1210 and both measures are based primarily on the same $^{10}\text{Be}_m$ measurements. (F) Log-log plot
 1211 adapted from von Blanckenburg et al. (2012) and updated comparing new mean $^{10}\text{Be}_i$ - and
 1212 $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ -derived sediment generation rates of the Potomac River (white square and black 1σ
 1213 standard deviation uncertainties; $^{10}\text{Be}_i$ sediment generation rate = $29.6 \pm 14.1 \text{ Mg km}^{-2} \text{ yr}^{-1}$;
 1214 $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ sediment generation rate = $40.0 \pm 21.7 \text{ Mg km}^{-2} \text{ yr}^{-1}$) with data derived from
 1215 sediment yields or $^{10}\text{Be}_i$ from other rivers throughout the Arctic (filled gray circles; Frank et al.,
 1216 2008), the Himalaya region (hollow gray circles; Rahaman et al., 2018), the northern United
 1217 States (filled black circles; Kusakabe et al., 1991), and South America (hollow black circles;
 1218 Brown et al., 1992; Wittmann et al., 2011, 2012, 2015). The $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ uncertainties are 50%
 1219 errors of the mean, or as reported in individual studies and $^{10}\text{Be}_i$ uncertainties are published
 1220 uncertainties of sediment generation rates. To plot data from Rahaman et al. (2017) and
 1221 Wittmann et al. (2015), denudation rates published in units of mm yr^{-1} were multiplied by the
 1222 published rock density ($2,600 \text{ kg m}^{-3}$). The uncertainties of $^{10}\text{Be}_m/^{9}\text{Be}_{\text{reac}}$ sediment generation
 1223 rates for all basins we sampled in the Potomac River basin represent a large amount of data
 1224 scatter, similar to estimated 50% uncertainties on means of other North American rivers. For
 1225 figure panels A-F, error bars as described above are shown; if error bars are not seen, they are
 1226 smaller than the panel symbology.

1227 Figure 4. Analyses of variance of $^{10}\text{Be}_i$ erosion rate data grouped and shaded by (A) land use and
 1228 (B) physiographic province. Box plot box is bounded by the 25th and 75th quartiles, and the
 1229 horizontal line in the box is the median; box plot whiskers encompass all data between the 25th
 1230 and 75th quartiles $\pm 1.5 \times$ the interquartile range. Samples that fall outside the statistical range of
 1231 the subsets are keyed to the dominant land use or by the physiographic province they are in. $^{10}\text{Be}_i$
 1232 erosion rate means are indistinguishable when grouped by land use (p values in panel A); when
 1233 grouped by physiographic province, $^{10}\text{Be}_i$ erosion rate means are statistically indistinguishable,
 1234 though right at the threshold of being different (p value in panel B), likely due to the lower

1235 erosion rates in the Coastal Plain.
1236 Figure 5. Log-log plot of ${}^9\text{Be}_{\text{reac}}$ -normalized ${}^{10}\text{Be}_{\text{m}}$ concentrations shows no statistical inverse
1237 relationship to ${}^{10}\text{Be}_{\text{i}}$ -derived sediment flux within the Potomac River basin as a whole, or by
1238 physiographic province.
1239 Figure 6. Erosion indices (EIs) of sampled basins for which there are multiple decades of
1240 sediment yield data (Gellis et al., 2004) show more ${}^{10}\text{Be}_{\text{m}}$ export from headwater basins in the
1241 Appalachian Plateau (AP) and Valley and Ridge (V&R) and from the Piedmont (PIED), where
1242 agricultural land use is intensive. Individual panels (A–C) remove catchment overlap for
1243 visibility. EIs for each sampling site are valid for the entire upstream basin area. EI values are
1244 given in parentheses after each sample ID. Data from Brown et al. (1998) are indicated by U.S.
1245 Geological Survey (USGS) stream gauging station IDs. The Hybla Valley core site is indicated
1246 by the white star in panel A. The remainder of map symbology is the same as in Figure 1. BR—
1247 Blue Ridge, CP—Coastal Plain.
1248 Figure 7. Mean $\pm 1\sigma$ standard deviations of ${}^{10}\text{Be}_{\text{i}}$ -derived erosion rates for outcrops in the upper
1249 reaches of the Potomac River basin compared to basin-wide ${}^{10}\text{Be}_{\text{i}}$ erosion rates. Differences
1250 between erosion rates in the Appalachian Plateau provide evidence for relief growth over 10^4 yr
1251 timescales, but not in the Valley and Ridge or Blue Ridge Provinces, where outcrop and basin-
1252 averaged erosion rates overlap within 1σ uncertainties. Bedrock data for the Appalachian Plateau
1253 come from Hancock and Kirwan (2007), Portenga et al. (2013), and Reuter (2005); bedrock data
1254 for the Valley and Ridge come from Duxbury et al. (2015) and Portenga et al. (2013); bedrock
1255 data for the Blue Ridge come from Duxbury et al. (2015), Hancock et al. (2015), and Portenga et
1256 al. (2013). Basin-averaged data for the Appalachian Plateau come from this study; basin-
1257 averaged data for the Valley and Ridge and Blue Ridge come from this study and Duxbury et al.
1258 (2015).
1259 Figure 8. ${}^{10}\text{Be}_{\text{m}}$ concentrations measured from samples taken from the Hybla Valley sediment
1260 core (circles) and from the modern-day Potomac River at POT01 (diamond), the furthest
1261 downstream, main river sample. Error bars on EIs are equal to the 1σ analytical uncertainty of
1262 the measured ${}^{10}\text{Be}_{\text{m}}$ concentrations, most of which are too small to be seen behind the data
1263 symbols. ${}^{10}\text{Be}_{\text{m}}$ measurements are connected by dashed black line for visibility only; the dashed
1264 black line does not indicate data continuity between samples. Results indicate that there is more
1265 erosion of topsoils, which are rich in ${}^{10}\text{Be}_{\text{m}}$, during colder climate periods and less erosion –
1266 lower ${}^{10}\text{Be}_{\text{m}}$ concentrations – during warmer climate periods. The age-depth relationship for the
1267 Hybla Valley core comes from Litwin et al. (2013). Solid gray line shows oak species abundance
1268 (*Quercus* spp.) and its changes through time, with decreases in abundance indicating colder
1269 climatic periods (Litwin et al., 2013).

Figure 1



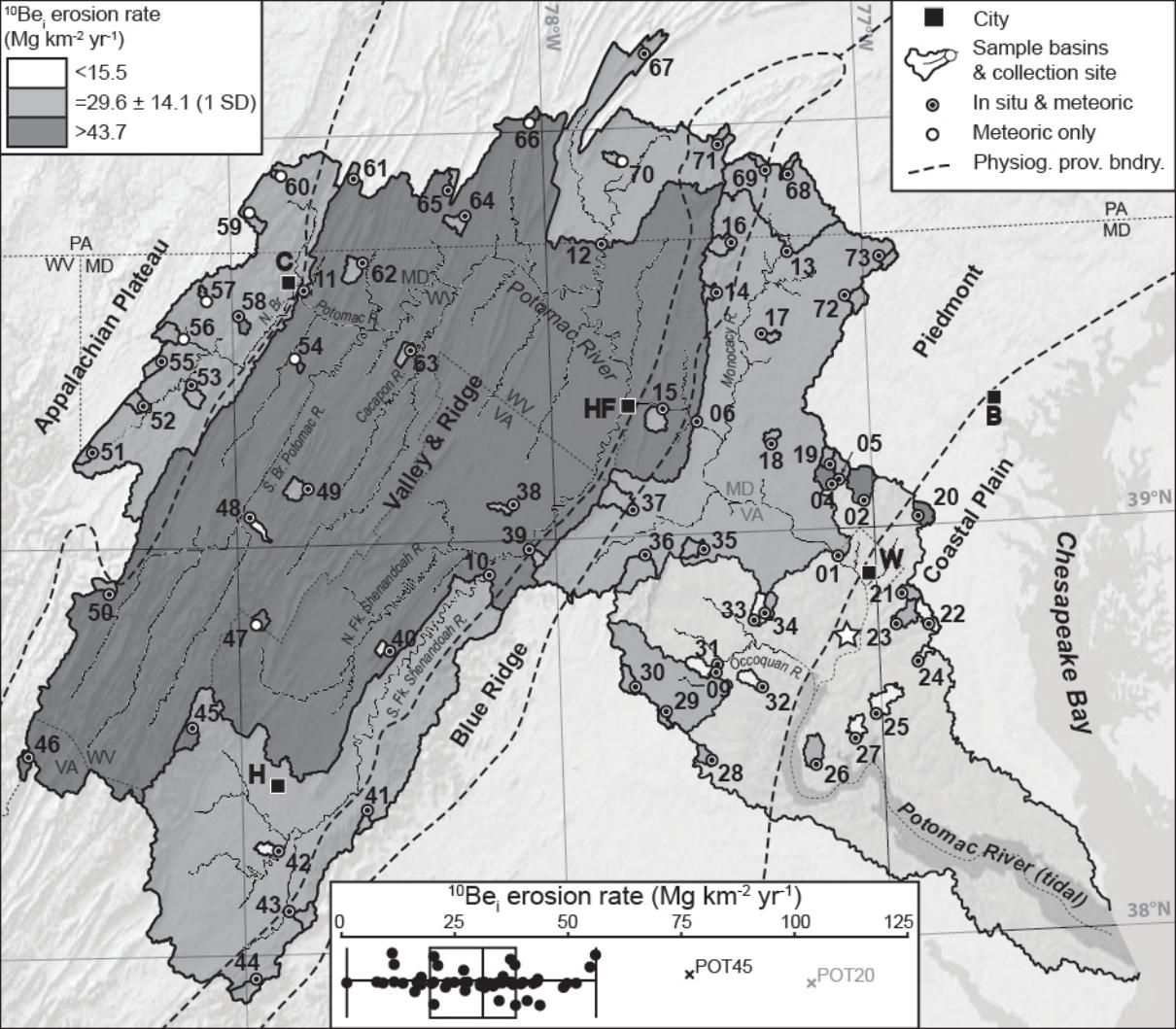
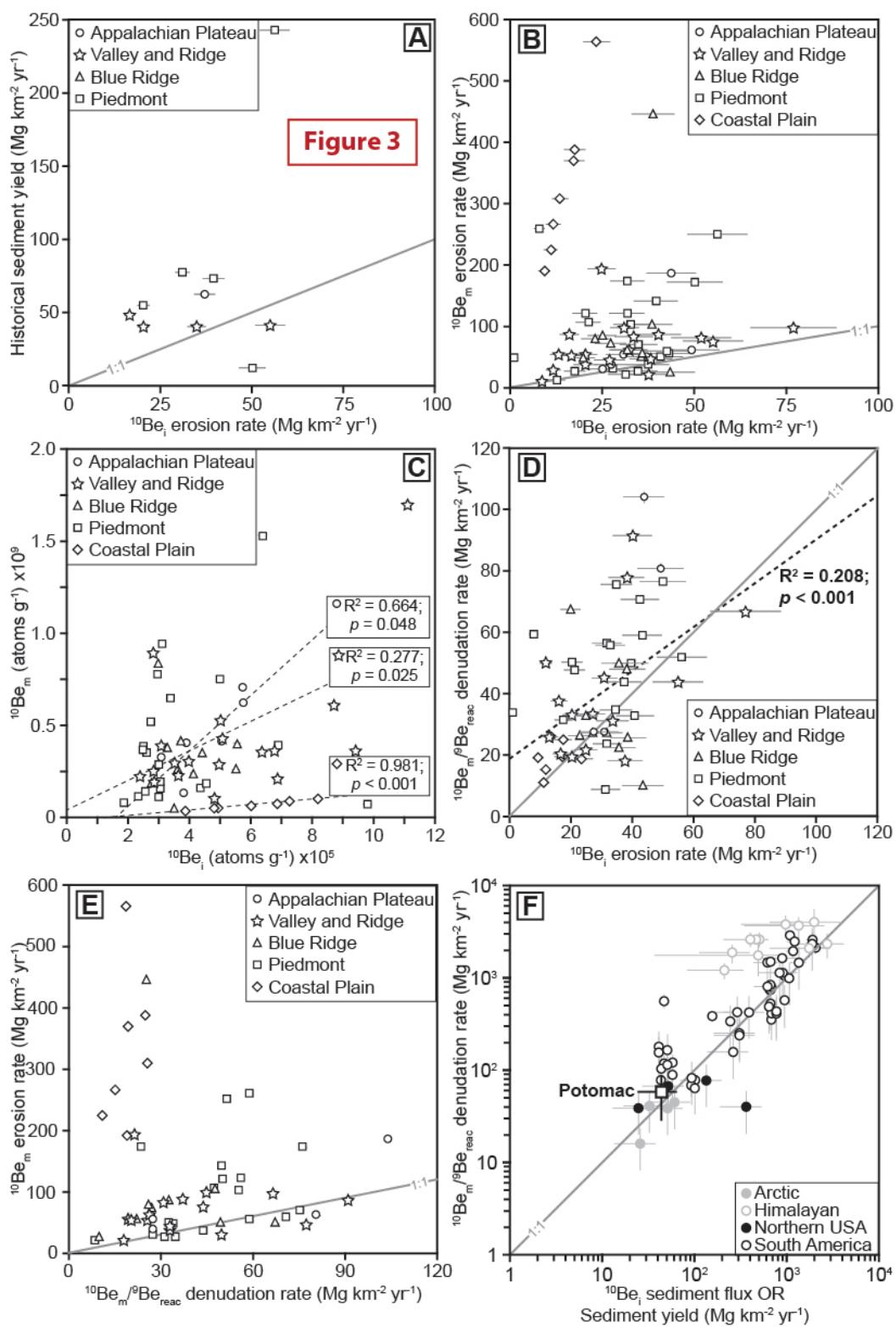
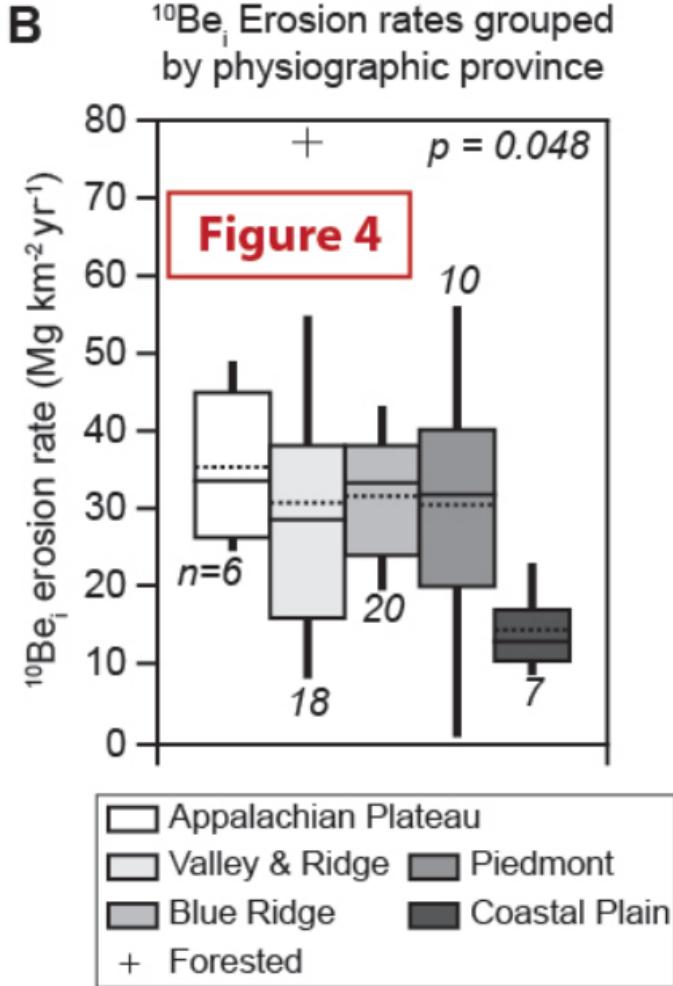
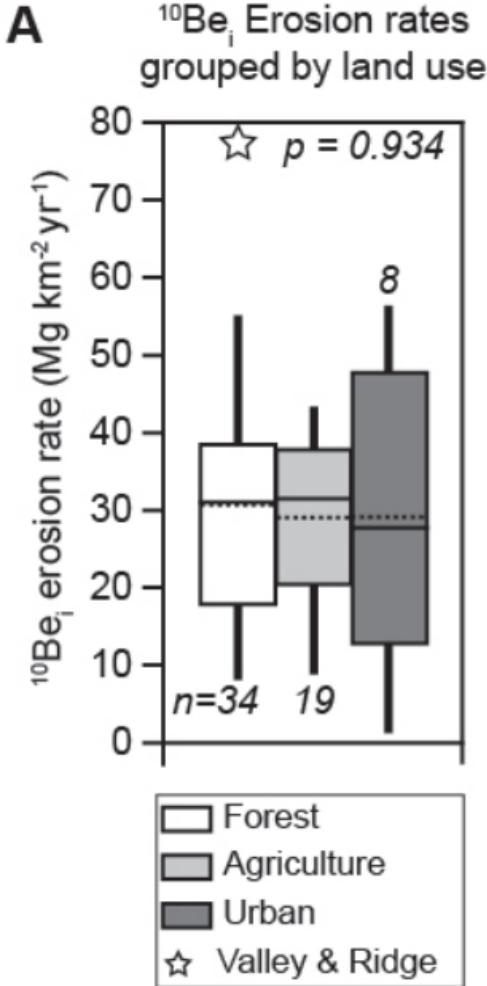


Figure 3





10^{-7}

Figure 5

Appalachian Plateau \circ

Valley & Ridge \star

Blue Ridge \triangle

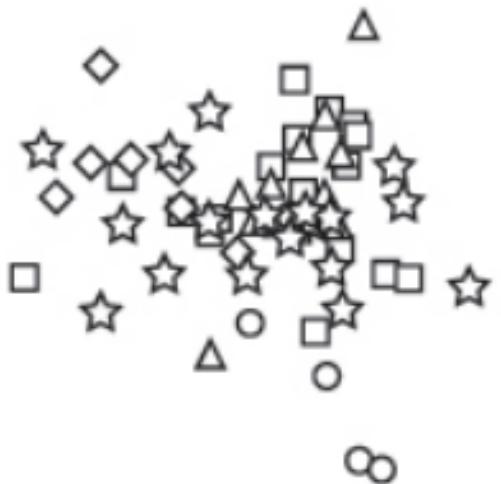
Piedmont \square

Coastal Plain \diamond

$^{10}\text{Be}_m / ^9\text{Be}_{\text{react}}$

 10^{-8} 10^{-9} 1 10 10^2 10^3

$^{10}\text{Be}_i$ erosion rate ($\text{Mg km}^{-2} \text{yr}^{-1}$)



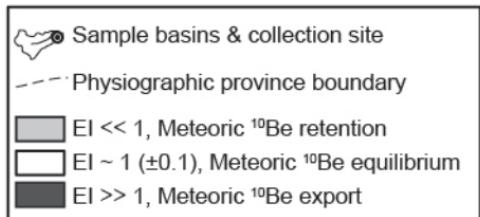
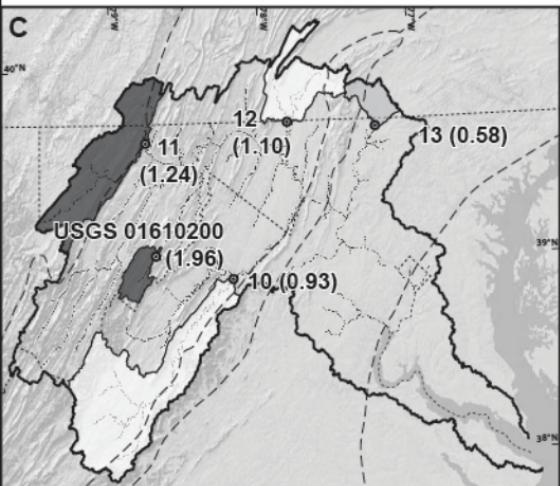
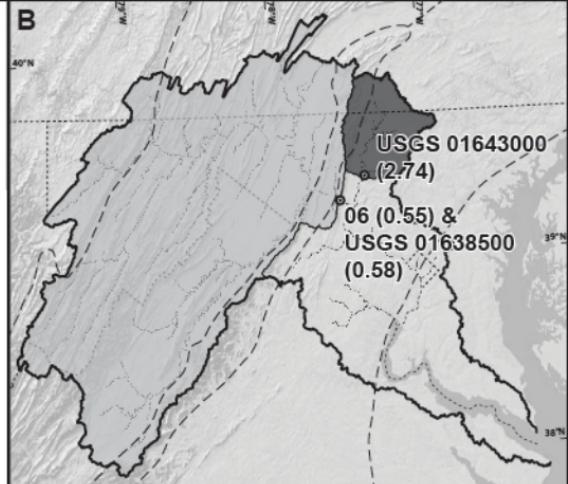
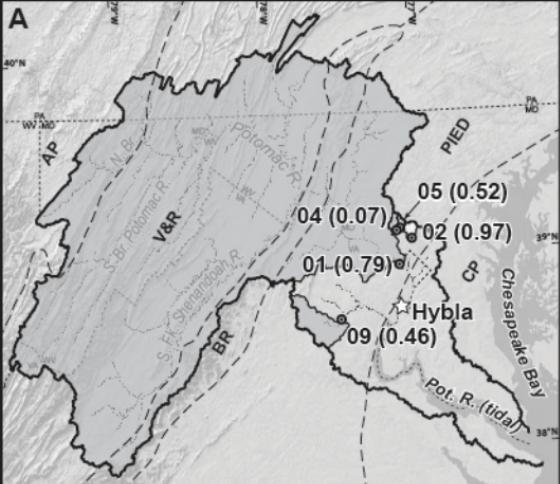
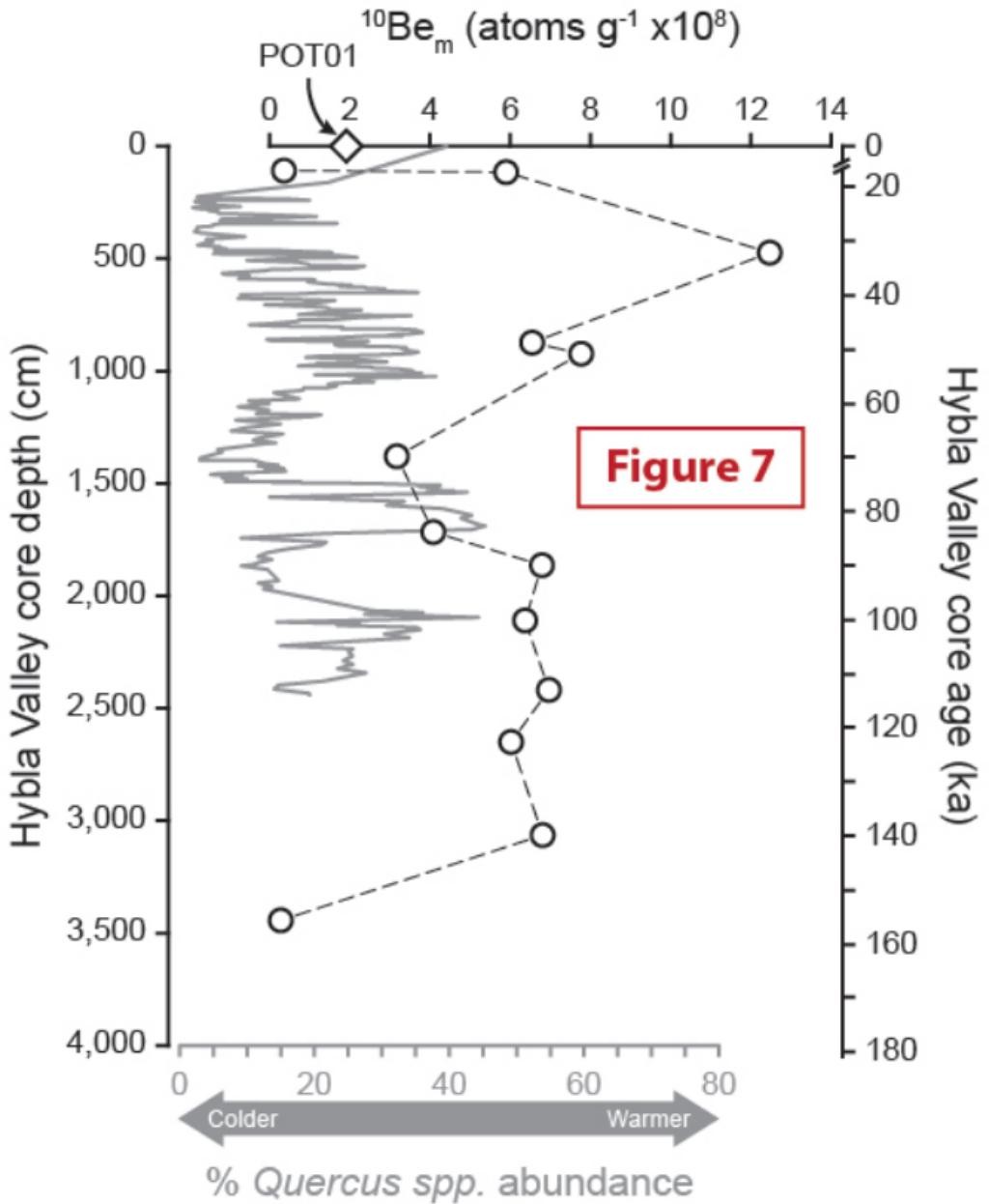


Figure 6



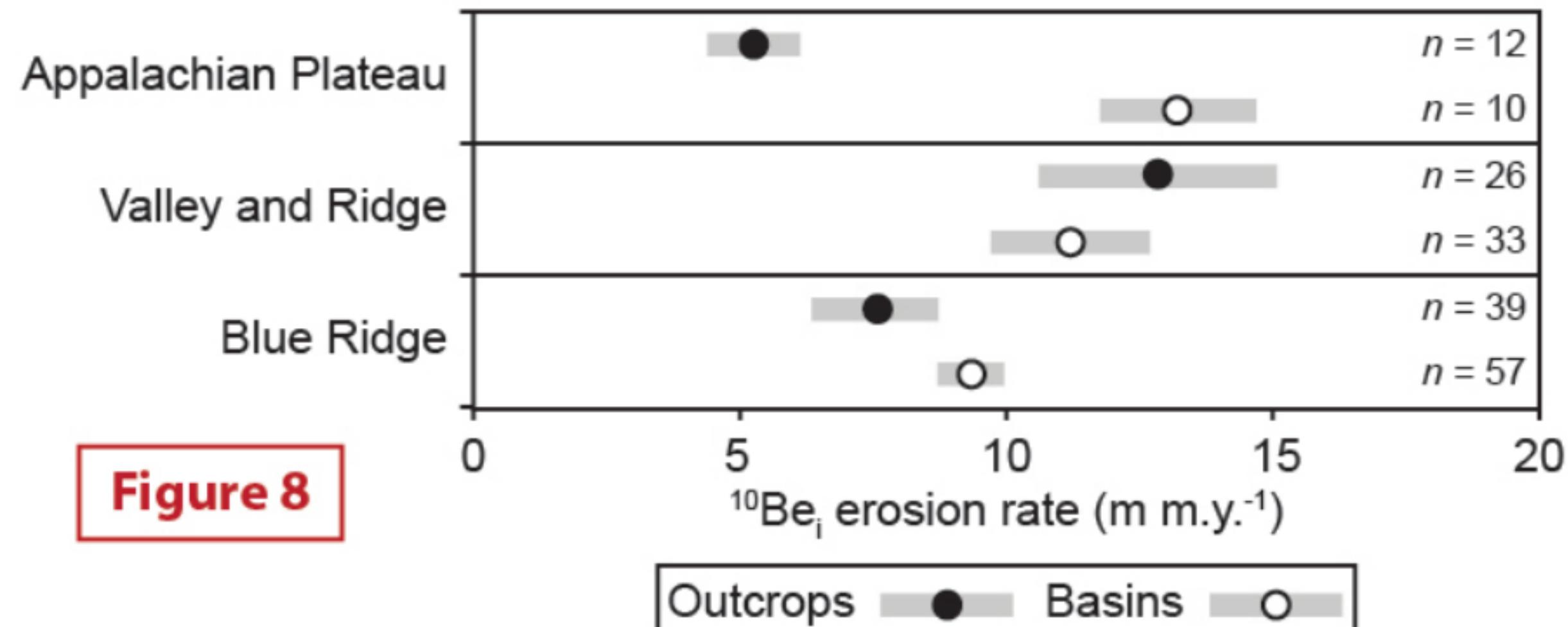


Figure 8