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Cosmogenic (un-)steadiness revealed by paired-nuclide catchment-wide denudation rates in the formerly half-glaciated Vosges Mountains (NE France)

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Abstract

Although catchment-wide denudation rates inferred from *in situ* cosmogenic nuclide concentrations measured in stream sediments has represented a ground-breaking progress in geomorphology over the last three decades, most of these studies rely on ^{10}Be concentrations only. It seems that this current and routine one-nuclide approach to infer catchment-wide denudation rates has somehow overshadowed two key assumptions that are cosmogenic steady-state and short sediment transit time at the catchment scale. Although a paired-nuclide approach allow testing these assumptions, it is rarely performed on stream sediments and this can become highly problematic in slow-eroding, formerly glaciated contexts. In this study, we thus measure both ^{10}Be and ^{26}Al in stream sediments pertaining to twenty-one rivers draining an entire low mountain range: the Vosges Massif (NE France). The latter exhibits a sharp gradient between its southern and northern part in terms of lithology, morphometry and climate. Moreover, if its northern part remained void of glacial cover during Quaternary cold stages, its southern part was significantly and repeatedly glaciated. We aim to assess the factors that control the denudation of the Vosges Mountains and to quantitatively explore the impact of both repeated glacial cover and storage of glacially derived sediments on $^{26}\text{Al}/^{10}\text{Be}$ ratios, hence cosmogenic (un-)steadiness in modern river samples. Our results first show that elevation, slope, channel steepness and precipitation are primarily organised along a N-S increasing trend. ^{10}Be - and ^{26}Al -derived catchment-wide denudation rates accordingly range from 34 ± 1 to 66 ± 2 , and 41 ± 3 to 73 ± 7 mm/ka, respectively, in thirteen investigated catchments that are in cosmogenic equilibrium. Lithological contrasts

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3 29 may control the pattern of denudation with a higher erodibility of the sandstone-dominated catchment to the north
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5 30 compared to the crystalline-dominated catchments to the south.
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7 31 Our results also show that catchments in strong cosmogenic disequilibrium ($^{26}\text{Al}/^{10}\text{Be}$ ratios from 1.4 to 5.2)
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9 32 spatially cluster in the SW part of the Vosges Mountains that was the most intensively glaciated during Quaternary
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11 33 cold stages. If this precludes any conclusion about controlling factors at the whole massif scale, this study is the
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13 34 first to quantify the impact of past glaciations on cosmogenic (un-)steadiness measured in stream sediments. A
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15 35 statistically significant relationship between the degree of depletion of the $^{26}\text{Al}/^{10}\text{Be}$ ratios and the spatial pattern
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17 36 of glaciation is found: the larger the former glacial cover in each catchment, the lower the $^{26}\text{Al}/^{10}\text{Be}$ ratio. Equally
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19 37 important is the significant correlation reported between the degree of depletion of the $^{26}\text{Al}/^{10}\text{Be}$ ratios and the
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21 38 proportion of glacial and fluvio-glacial deposits within each catchment. These two relationships underline the link
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23 39 between cosmogenic unsteadiness in the stream cosmogenic signal and long-lasting and repetitive ice shielding,
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25 40 and complex sediment routing systems in glacial environments, respectively. We thus argue to systematically
26
27 41 measure ^{26}Al in complement to ^{10}Be and to test the steady-state assumption when it comes to infer catchment-wide
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29 42 denudation rates from modern stream sediments, especially in slow eroding, formerly glaciated landscapes.
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33 34 43 35 36 44 **1. Introduction**

37
38 45 Assessment of catchment-wide denudation (CWD) rates inferred from *in situ* cosmogenic nuclide concentrations
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40 46 measured in modern stream sediments has represented a ground-breaking progress in geomorphology over the last
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42 47 three decades (von Blanckenburg, 2005; Dunai, 2010; Granger and Schaller, 2014). This approach has been
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44 48 applied to fluvial catchments from practically every tectonic and climatic setting worldwide, including currently
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46 49 (e.g. Grin et al., 2018) and formerly (e.g. Buechi et al., 2014) glaciated terrains. The key research aim of those
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48 50 studies consists in disentangling the complex interplays between topography (e.g. Peifer et al., 2021), tectonics
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50 51 (e.g. Olivetti et al., 2016), climate (e.g. Carretier et al., 2013) and lithology (e.g. Cyr et al., 2014) which primarily
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52 52 control denudation. The thousands of river-sediment samples collected in the majority of case studies, however,
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54 53 were processed to measure *in situ* ^{10}Be concentrations only (Portenga and Bierman, 2011). Although this single-
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56 54 nuclide approach relies on several key assumptions, it seems that the current ^{10}Be 's routine approach used to infer
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58 55 CWD rates has somehow overshadowed two of them. First, little attention has been recently paid to cosmogenic
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3 56 steady-state at the catchment-scale (von Blanckenburg, 2005). It implies that the catchment's inbound flux via
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5 57 ^{10}Be production is counterbalanced by the outbound flux by denudation and decay at the catchment's outlet where
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7 58 the sampling takes place. Second, the timespan over which sediments are transported within the catchment to the
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9 59 outlet, i.e. sediment transit time, must be short compared to the timespan over which denudation occurs (Dunai,
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11 60 2010). In settings where these conditions might not be met, ^{10}Be radioactive decay can become sizeable so that
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13 61 special care must be taken in settings where long-term sediment storage and remobilisation is evidenced. Neither
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15 62 the assumption of catchment's steady-state nor that of short sediment transit time can be checked by processing
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17 63 river-sand ^{10}Be concentrations only. As for steady-state, glacier cover, glacial erosion and/or glacially derived
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19 64 sediments may significantly impact the estimation of ^{10}Be -derived CWD rates in currently (e.g. Delunel et al.,
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21 65 2014) or formerly (e.g. Glotzbach et al., 2013) glaciated catchments. As for transit time, glacial and post-glacial
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23 66 environments have complex sediment routing systems (e.g. Porter et al., 2019) where reworking of ancient
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25 67 sediments impacts cosmogenic inventories as well (e.g. Wittmann et al., 2009; Buechi et al., 2014). A few ^{10}Be -
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27 68 based studies developed some modelling approaches to partially cope with this twofold key issue (Godard et al.,
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29 69 2012; Glotzbach et al., 2013).
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34 71 Examination of steady-state, transit time and potential storage are potentially better constrained by measuring both
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36 72 ^{10}Be and ^{26}Al concentrations in the same quartz-bearing sample (e.g. Bhattacharjee et al., 2023). Since its original
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38 73 formulation in Lal's (1991) paper, the two-nuclide diagram distinguishes those samples that meet the steady-state
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40 74 assumption by plotting in the so-called steady-erosion island (Dunai, 2010) from those that underwent complex
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42 75 exposure histories and are characterised by depleted $^{26}\text{Al}/^{10}\text{Be}$ ratios. This approach has been widely used in
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44 76 formerly glaciated regions of e.g. Greenland (Corbett et al., 2013; Strunk et al., 2017; Andersen et al., 2020) and
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46 77 Scandinavia (Andersen et al., 2018; Knudsen and Egholm, 2018) to successfully assess glacial erosion efficiency
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48 78 and denudation rates. Depleted $^{26}\text{Al}/^{10}\text{Be}$ ratios in many glacial bedrock and erratic samples clearly violate the
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50 79 steady-state assumption due to repeated and extensive burial under thick ice masses with low erosive potential,
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52 80 i.e. cold-based glaciers. On the contrary, the paired-nuclide ($^{26}\text{Al}-^{10}\text{Be}$) approach was only applied in few studies
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54 81 for modern stream sediments, although Wittmann et al. (2020) recently highlighted significantly depleted $^{26}\text{Al}/^{10}\text{Be}$
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56 82 ratios in 35% of the >50 large rivers they studied all over the Earth's surface. Merely a handful of these studies
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3 83 dealt with glaciated catchments (Safran et al., 2005; Hippe et al., 2012; Grin et al., 2018; Wittmann et al., 2020;
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5 84 Zhang et al., 2021), but cosmogenic disequilibrium is highlighted not only in currently glaciated catchments of
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7 85 Tibet (Zhang et al., 2021) but also in formerly and marginally glaciated catchments of the Bolivian Andes (Hippe
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9 86 et al., 2012). However, Zhang et al. (2021) could only conclude that sediment reworking is a potential issue without
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11 87 further exploration nor quantification on this process. This altogether demonstrates our current lack of
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13 88 understanding in how cosmogenic signals measured in modern stream sediments might be perturbed in (formerly)
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15 89 glaciated catchments.
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21 91 Located in north-eastern France, the Vosges Massif (or Vosges Mountains; VM) is a favourable setting to
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23 92 investigate this question. This S-N oriented low-elevation mountain range exhibits a sharp dissimilarity between
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25 93 its southern and northern part in terms of lithology, morphometry and climate. Topographic imprints from
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27 94 successive Quaternary glaciations in the VM landscape also follow this spatial pattern: whereas most of the
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29 95 crystalline southern VM were repeatedly covered and shaped by thick glaciers (Flageollet, 2002; Mercier, 2014),
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31 96 the northern half was void of ice cover (Darmoiois-Théobald, 1972; Heyman et al., 2013). Taking advantage of the
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33 97 VM setting and characteristics, this study aims at (i) demonstrating the spatial structuration of the VM in terms of
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35 98 first-order controlling factors for catchment-scale denudation, (ii) evaluating the cosmogenic (dis)equilibrium in
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37 99 the VM and (iii) quantitatively exploring the impact of both repeated glacial cover and storage of glacially derived
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39 100 sediments on paired cosmogenic ratios. First, morphometric and climatic indices are computed and groups of
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41 101 lithologically uniform catchments are statistically determined. Second, modern stream sediments from 21 river-
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43 102 catchments outlets draining the whole VM are sampled for *in situ* ^{10}Be and ^{26}Al concentration measurements.
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45 103 Third, a systematic comparison is undertaken between $^{26}\text{Al}/^{10}\text{Be}$ ratios and high-resolution data including previous
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47 104 glacial coverage, as well as the spatial distribution of glacial and fluvio-glacial sediments.
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52 105 53 54 106 **2. Study area** 55

56 107 The VM form the western footwall block of the southern Upper Rhine Graben (north-eastern France; Fig. 1A). Its
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58 108 uplift is assigned to Miocene lithospheric folding resulting from NW-directed compressional stresses exerted in
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60 109 the Alpine foreland (e.g. Ziegler and Dèzes, 2007). Plio-Quaternary tectonic re-activation and uplift were assumed
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3 110 as well (Harmand and Cordier, 2012). The VM's lithology primarily is bipartite: Paleozoic crystalline rocks and
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5 111 Triassic sandstones compose the southern (~2/3) and northern (~1/3) parts, respectively (Fig. 1B). Strongly
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7 112 contrasting with the almost uniform Mesozoic sedimentary cover to the north, the Paleozoic basement reaches
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9 113 Hercynian orogenic units with various lithologies (Fig. 1B; Skrzypek, 2011). Plutonic rocks prevail (i.e. granites
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12 114 and (grano-)diorites mostly) but metamorphic (i.e. mostly gneiss) and volcanic (i.e. various lavas and tuffs) rocks
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14 115 also occur.

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16 116 The VM exhibit a well-marked N-S topographic gradient, from a low- to medium-elevation mountain range from
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18 117 north to south. Mean and peak elevations along three WNW-ESE oriented swath profiles performed in the VM's
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21 118 northern, central and southern parts amount to ~255/525, 470/1090 and 710/1350 m asl, respectively (Fig. 2A). A
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23 119 southward massif-scale increasing trend in normalised channel steepness index reflects this gradient as well (Fig.
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25 120 1C). The VM is sharply bounded by the Vosgian normal fault system bordering the Upper Rhine Graben whereas
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27 121 its western side falls upon the slightly tilted Mesozoic formations from the Paris Basin (Figs. 1, 2A). Owing to the
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30 122 NNE/SSW-orientation of its main topographic divide, the VM typically form an almost perpendicular orogenic
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32 123 barrier to the predominant oceanic western atmospheric flow, thereby inducing a typical rain shadow effect (Fig.
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34 124 1D; François and Humbert, 2000). Mean annual rainfall concomitantly increases with elevation along the main
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36 125 ridge as well, ranging from about 900 mm/a in the northern part to up to 1400 mm/a at elevations higher than 1350
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38 126 m (Fig. 1D). Deciduous, resinous and mixed forests cover no less than ~70% of the VM's area (Rochel, 2004).
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41 127 They essentially occur on hillslopes and plateau-top positions, regardless the geographic location within the massif
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43 128 (except the bare, highest interfluves in the southern part; Fig. 2Bc).

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47 130 The spatial pattern of Quaternary glaciations and their imprints on the landscape reflect this massif-scale
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50 131 dichotomy (Fig. 1E; Flageollet, 2002; Mercier & Jeser, 2004; Mercier, 2014). To the north, roughly two thirds of
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52 132 the VM exhibit fluvially overprinted landscapes (Fig. 2Ba-b). They completely lack evidence of former glacial
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54 133 activity and allow inferring an ice-free area in the northern third of the VM (Flageollet, 2002). Fluvial landscapes
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56 134 of the central part bear witness of local and sparse evidence of low-magnitude past glacial activity, i.e. small-
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58 135 dimensioned cirques and cirque moraines (Darmois-Théobald, 1972; Fig. 1E). Whilst mean fluvial incision usually
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60 136 ranges from ~150 (Fig. 2Ba) to 300 m (Fig. 2Bb) in the northern and central third, respectively, this whole area

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3 137 most probably experienced periglacial conditions during Quaternary cold periods. By contrast, the VM's southern
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5 138 third (with mean elevations >700 m) exhibit glacially overprinted landscapes. Numerous well-developed cirques
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7 139 along the main topographic divide, U-shaped valleys, glacial deposits (e.g. moraines; Fig. 2Bc), along with glacial
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9 140 and fluvio-glacial deposits blanketing the valley floors (Supplementary Material, Fig. A1) all indicate large-scale,
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11 141 past glacial activity. Despite the lack of numerical dating on glacial landforms, these landscapes bear witness of
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13 142 several Quaternary glaciations characterised by various ice extents (Fig. 1E). Flageollet (2002) recognised at least
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15 143 three to five glaciations. Spatial extensions of the supposedly Last Glacial Maximum (Marine Isotope Stage –
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17 144 MIS2) and the penultimate glaciation (MIS6) are judged reliable because they are based on abundant field
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19 145 evidence, contrasting with the poorly constrained pre-MIS6 glaciations (Supplementary Material, Fig. A2).
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21 146 However, the study of sedimentary formations located at the VM's south(west-)ern margin lead Ménillet (2019)
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23 147 to consider the occurrence of Vosgian glaciations during the Early Pleistocene whose remains were erased during
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25 148 subsequent cold episodes. We thus suggest that the VM has repeatedly experienced significant glaciations over
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27 149 the whole Quaternary.
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32 150 33 34 151 **3. Data and methodology**

35 36 152 **3.1. Lithology, morphometry and climate**

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38 153 Massif-scale lithology was characterised using the 1:50.000 vectorised geological map provided by the French
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40 154 Geological Survey (BRGM; [https://infoterre.brgm.fr/formulaire/telechargement-cartes-geologiques-](https://infoterre.brgm.fr/formulaire/telechargement-cartes-geologiques-departementales-150-000-bd-charm-50)
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42 155 [departementales-150-000-bd-charm-50](https://infoterre.brgm.fr/formulaire/telechargement-cartes-geologiques-departementales-150-000-bd-charm-50), last access 29th January 2021). We performed a query-attribute
43
44 156 reclassification to obtain five main lithologies for bedrock units: plutonic, metamorphic, volcanic and two classes
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46 157 in sedimentary rocks, i.e. sandstone (mostly Triassic sandstones) and marno-calcareous (at the VM's margins).
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48 158 Quaternary deposits were treated separately and likewise subdivided into alluvial, colluvial and glacial/fluvio-
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50 159 glacial sediments. The respective proportions of each rock and deposit class were extracted within each sampled
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52 160 catchment (Fig. 3). Hierarchical clustering analysis was applied to our dataset of 21 catchments based on the
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54 161 aforementioned lithological classes. Further details on the method is given in Supplementary Material (part 1.1).
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56 162 We used the 25 m-resolution digital elevation model (DEM) provided by the French National Institute of
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58 163 Geography (BD ALTI) to compute three commonly used catchment-averaged metrics: elevation, slope and
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3 164 channel steepness index. For each DEM pixel, slope was computed as the steepest gradient between the eight
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5 165 neighbouring pixels, using the function *terrain* of the *raster* package in R (Hijmans, 2022). Normalised channel
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7 166 steepness index (k_{sn}) was computed using the LSD Topotools software (Mudd et al., 2018) with a reference
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9 167 concavity of 0.45 for inter-catchment comparison (Fig. 1C). We finally computed the mean annual precipitation
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12 168 over the VM (8-km resolution; Fig. 1D) from monthly-cumulated precipitation over the 1958-2009 period
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14 169 extracted from the national hydroclimatic database of Météo-France (Vidal et al., 2010). Catchment-averaged
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16 170 values for each topographic and climatic metric (i.e. elevation, slope, channel steepness, precipitation) were finally
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18 171 computed within each studied catchment (Supplementary Material Table S1).
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20 21 172 22 23 173 **3.2. Sampling strategy, AMS measurements and denudation rates**

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25 174 We sampled 22 stream sediments from 21 distinct rivers (two samples for the Fecht River; Table 1). Our sampling
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27 175 strategy covers all main rivers draining the VM along with some of their intra-massif tributaries (Fig. 1). Individual
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29 176 catchment sizes range from 62 to 1037 km² and the total surface considered in this study represents ~4,650 km²
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32 177 (Fig. 1). Sampling sites were selected at the outlet of the rivers' mountainous reach. Buechi et al.'s (2014) sampling
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34 178 strategy, primarily focusing on fluvially overprinted landscapes over glacially overprinted landscapes, was adapted
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36 179 with slight modifications (Figs. 1E, 2): the northern and southern parts of the sampled catchments overall match
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38 180 the fluvial landscapes and the glacial and fluvio-glacial landscapes, respectively.
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43 182 River-sand samples were dried and sieved at the 250-1000 μm grain-size fraction. All ²⁶Al and ¹⁰Be chemical
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45 183 extractions and were performed at LN2C (National Laboratory of Cosmogenic Nuclides) located at the Centre
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47 184 Européen de Recherche et d'Enseignement des Géosciences de l'Environnement (CEREGE) laboratory in Aix-
48
49 185 en-Provence (France) following standard procedures (e.g. Braucher et al., 2011; see Supplementary Material part
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51 186 1.2 for further details). AMS measurements took place at the French national Facility, ASTER (Arnold et al.,
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53 187 2010). Measurement calibrations of the beryllium and aluminium measurements were done using the STD-11
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55 188 standard (¹⁰Be/⁹Be ratio of $1.191 \times 10^{-11} \pm 1.08\%$; Braucher et al., 2015) and the ASTER in-house standard SM-
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57 189 AL-11 (²⁶Al/²⁷Al ratio of $7.401 \pm 0.064 \times 10^{-12}$; Merchel & Bremser, 2004), respectively. The reported analytical
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59 190 uncertainties (1σ) include both the AMS counting statistics and internal error (0.5% for ¹⁰Be; Arnold et al., 2010),
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3 191 chemical blank measurement and ^{27}Al measurements. $^{10}\text{Be}/^9\text{Be}$ blank ratio was 2.15×10^{-15} and $^{26}\text{Al}/^{27}\text{Al}$ blank
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5 192 ratio was less than 7.02×10^{-16} .
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9 194 CWD rates were inferred from ^{10}Be and ^{26}Al concentrations using the software CosmoCalc version 3.1
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11 195 (<https://www.ucl.ac.uk/~ucfbpve/cosmocalc/>; Vermeesch, 2007), based on the time-independent Lal (1991)/Stone
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13 196 (2000) scaling model. The 22 catchments were first delineated using the 25-m resolution DEM and standard
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15 197 hydrological routing procedures. Latitude of each catchment's centroid as well as their average elevation were
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17 198 then calculated. For the sea-level-high-latitude (SLHL) reference production rates of ^{10}Be and ^{26}Al , we used the
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19 199 default values implemented in CosmoCalc version 3.1 which are 4.3 and 30.95 at/g/a, respectively. Muonic
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21 200 productions was based on the scheme of Braucher et al. (2011). We assumed a standard bedrock density of 2.7
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23 201 g/cm³ and did not apply any production correction for topographic shielding as reported by DiBiase (2018), nor
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25 202 for snow shielding.
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32 204 **4. Results**

33 205 **4.1. Lithological, morphometric and climatic spatial patterns**

34 206 The spatial distribution of the different bedrock lithologies and Quaternary deposits reveals significant spatial
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36 207 variations over the VM and the 22 sampled catchments (Fig. 3). Some catchments are clearly dominated by a
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38 208 single main lithology, such as plutonic (e.g. #13, 20) and metamorphic (e.g. #7) bedrocks or sandstone (e.g. #10,
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40 209 15). By contrast, other catchments are either constituted of two (e.g. #5, 14) or more (e.g. #1, 19) dominant
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42 210 lithologies. The hierarchical clustering analysis results in four catchments classes that are named after their main
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44 211 respective lithological composition: metamorphic (m), plutonic (p), sedimentary (s) and volcanic (v). The relative
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46 212 proportions of Quaternary deposits in each catchment not only significantly vary, ranging from 4.5% (#6) to 48.3%
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48 213 (#2), but the prevailing type of Quaternary deposits also differs with dominant alluvial, colluvial and glacial/fluvio-
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50 214 glacial sediments in e.g. catchments #16, 11 and 14, respectively (Fig. 3). Further details on the lithological
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52 215 specificities of each group of catchments are given in Supplementary Material (part 2.1).
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3 217 Latitudinal variations in catchment-averaged elevation, slope, k_{sn} and annual precipitation range from 260 to 862
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5 218 m, 8 to 21°, 8 to 69 m^{0.9} and 748 to 1767 mm, respectively (Table 1 and Fig. 4). Whereas elevation, slope and
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7 219 annual precipitation vary by a factor of 2 to 3 at the massif scale, a southward increasing trend in k_{sn} of almost one
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9 220 order of magnitude is documented. Mean elevation (Fig. 4A), slope (Fig. 4B), k_{sn} (Fig. 4C) and annual precipitation
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11 221 (Fig. 4D) all display a significant inverse correlation with the catchment-centroid latitude ($R^2=0.42-0.81$) and
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13 222 thereby attest the VM's N-S gradient. A secondary W-E gradient is also observed for topographic and climatic
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15 223 metrics, with two diverging trends. Unlike slope and k_{sn} values that are higher in the east than in the west (Fig.
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17 224 4B-C), higher annual precipitation are recorded in the west (Fig. 4D). No significant W-E difference is found
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19 225 concerning the elevation (Fig. 4A).
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23 226 24 25 227 **4.2. ¹⁰Be and ²⁶Al concentrations and ²⁶Al/¹⁰Be ratios**

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27 228 Measured ¹⁰Be and ²⁶Al concentrations, calculated ²⁶Al/¹⁰Be ratios and associated uncertainties are presented in
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29 229 Table 1. ¹⁰Be and ²⁶Al concentrations range from 6.1(±0.2) to 13.7(±0.5) x 10⁴ at/g and from 11.1(±2) to 92.5(±9)
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31 230 x 10⁴ at/g, respectively. ²⁶Al/¹⁰Be ratios for the 22 samples range from 1.4 ±0.2 to 8.0 ±0.7. ²⁶Al/¹⁰Be ratios are
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33 231 plotted in the two-nuclide diagram (Lal, 1991; Fig. 5A) using CosmoCalc version 3.1 (Vermeesch, 2007) with the
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35 232 time-independent Lal (1991)/Stone (2000) scaling model. In this diagram, the steady-erosion island is comprised
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37 233 between two lines representing the evolution of the ²⁶Al/¹⁰Be ratio in material being continuously exposed without
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39 234 erosion (upper line) and varying steady erosion rates (lower line). Thirteen samples (#1, 4, 5, 7, 8, 9, 10, 11, 15,
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41 235 16, 17, 20, 21) out of 22 modern stream samples (~60%) plot within the steady-erosion island, considering 2σ
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43 236 uncertainties (Fig. 5A). They are referred to as the *steady-state samples* hereinafter. The remaining nine samples
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45 237 (#2, 3, 6, 12, 13, 14, 18, 19, 22) plot below the steady-erosion island, considering 2σ uncertainties (Fig. 5A). In
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47 238 the so-called complex exposure zone, these samples show different apparent burial durations (i.e. minimum burial
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49 239 times) due to their varying depleted ²⁶Al/¹⁰Be ratios (Fig. 5A): these range from ~500 ka (#6, 22) to more than 3
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51 240 Ma (#2, 14; Table 1). They are referred to as the *non-steady-state samples* hereinafter.
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55 241 56 57 242 **4.3. Catchment-averaged denudation rates**

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3 243 We consider here the whole dataset regardless of the individual $^{26}\text{Al}/^{10}\text{Be}$ ratios (Table 1 and Fig. 5B). CWD rates
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5 244 and integration times inferred from ^{10}Be concentrations range from 34 ± 1 to 88 ± 4 mm/ka (catchments #11; 3) and
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7 245 6.8 to 17.6 ka, respectively. Those inferred from ^{26}Al concentrations range from 41 ± 3 to 363 ± 53 mm/ka
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9 246 (catchments #11; 2) and 1.7 to 14.5 ka, respectively (Table 1 and Fig. 5B). First, when comparing ^{10}Be and ^{26}Al -
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11 247 inferred CWD rates (Fig. 5B), the *steady-state samples* are well distributed along the 1:1 line ($\pm 25\%$). By contrast,
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13 248 among the nine *non-steady-state samples*, seven of them (#2, 3, 12, 13, 14, 18, 19) clearly plot apart from the 1:1
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15 249 line (Fig. 5B). Almost all ^{26}Al -inferred CWD rates are significantly higher than ^{10}Be -inferred values, sometimes
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17 250 by a factor four or higher (#2 and 14). Second, we plot the latitude of the catchments' centroid against CWD rates
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19 251 inferred from both nuclides, alongside the four lithological groups and the mean annual precipitation (Fig. 6). On
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21 252 the eastern side, a southward increasing trend is highlighted for both ^{10}Be and ^{26}Al -derived CWD rates. On the
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23 253 western side, no clear spatial pattern can be observed, neither for ^{10}Be nor for ^{26}Al . Importantly, the seven *non-*
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25 254 *steady-state* samples showing the largest discrepancies in ^{10}Be - versus ^{26}Al -CWD rates are all located in the
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27 255 southern part of the VM (Fig. 6), regardless the catchment lithology and annual precipitation.
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34 257 **5. Discussion**

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36 258 **5.1. What can - and cannot - be said about spatial denudation patterns in the Vosges Mountains?**

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38 259 **5.1.1. Unsteadiness blurs cosmogenic nuclide signals at the massif scale**

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41 260 A global southward increasing trend in elevation, slope, k_{sn} and annual precipitation is highlighted throughout the
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43 261 VM together with a clear latitudinal distribution of the main bedrock lithologies (Figs. 3, 4). These have been
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45 262 proposed as the primary factors governing catchment-wide denudation based on ^{10}Be measured in active stream
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47 263 sediments (e.g. Safran et al., 2005; Carretier et al., 2013; Cyr et al., 2014; Mandal et al., 2015; Olen et al., 2016;
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49 264 Olivetti et al., 2016; Delunel et al., 2020; Peifer et al., 2021). Correlations are thus tested for elevation, slope, k_{sn}
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51 265 and annual precipitation (Fig. 7; Table A in Supplementary Material). Only one significant relationship (though
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53 266 weak: $R^2=0.22-0.24$; $p\text{-value}<0.03$) is observed between annual precipitation and CWD rates inferred from both
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55 267 nuclides and based on the whole VM dataset (Fig. 7B, E).

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59 268 One must recall, however, that nine catchments (#2, 3, 6, 12, 13, 14, 18, 19, 22; $\sim 40\%$) violate the steady-state
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61 269 assumption required to infer denudation rates from measurement of *in situ* cosmogenic nuclides concentrations in
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3 270 river sediments (von Blanckenburg, 2005; Dunai, 2010). Accordingly, these *non-steady-state samples* should be
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5 271 discarded when it comes to (i) infer CWD rates at the massif scale and (ii) correlate these with potential controlling
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7 272 factors (climatic and/or morphometric control). Discarding these catchments has a strong impact as they represent
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9 273 a total drainage area of ~1,987 km² (i.e. 43% of the study area; Fig. 1). Moreover, these catchments are mostly
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11 274 located in the VM's southern part (7/9) and thereby substantially reduce the relationship between the VM's N-S
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13 275 structuration and the spatial pattern of denudation (Fig. 7). Unsteadiness in cosmogenic nuclide concentrations
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15 276 measured in modern river sediments from nine catchments thus prevents any conclusions regarding the controlling
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17 277 factors at the whole massif scale.

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22 279 Two subsets can be distinguished among *non-steady-state samples*: the northern ones (# 6 and 22) have slightly
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24 280 depleted ²⁶Al/¹⁰Be ratios (5.7±0.5) whereas the southern ones (#2, 3, 12, 13, 14, 18, 19) have much lower ratios
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26 281 (1.4±0.2–5.2±0.6; Fig. 5A; Fig. 6). Whilst ¹⁰Be- and ²⁶Al-inferred CWD rates of the first group overlap the 25%
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28 282 uncertainty space around the 1:1 line defined by both nuclides (Fig. 5B), those of the second group systematically
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30 283 plot apart from it. We assume that the first two samples (# 6 and 22) might possibly fall into the range of
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32 284 uncertainties related to methodological effects on depletion of ²⁶Al/¹⁰Be production ratios (Wittmann et al., 2020).
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34 285 On the other hand, the strongly depleted ²⁶Al/¹⁰Be ratios peculiar to the second group cannot be explained by
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36 286 intrinsic uncertainties linked to the production ratio or attenuation lengths. One must consider instead the effect(s)
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38 287 of one or several geomorphological processes since no significant relationship is observed between catchment area
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40 288 and ²⁶Al/¹⁰Be ratios at the massif scale (Supplementary Material, Table A, Fig. C).

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45 290 *5.1.2. Lithological and vegetation potential control on denudation for steady-state catchments*
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47 291 With removing the nine *non-steady-state* samples, the range of ¹⁰Be- and ²⁶Al-derived CWD rates in the thirteen
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49 292 steady-state catchments is significantly reduced to 34±1 – 66±2 and 41±3 – 73±7 mm/ka, respectively. ¹⁰Be-
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51 293 derived CWD from a small unglaciated catchment located between our catchments #8 and 20 fall in the same
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53 294 range (i.e. ~38 mm/ka; Ackerer et al., 2022). By contrast, in these thirteen catchments, the observed large ranges
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55 295 in mean elevation, k_{sn} and annual precipitation remains as in the initial dataset (see 4.1) and only the slope
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57 296 fractionally decreases (between 8.3 and 20.4°; Table 1). Yet, none of the above morphometric and climatic

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3 297 controlling factors appears playing a major role here, as shown by the absence of significant correlation with CWD
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5 298 rates inferred from both nuclides (Supplementary Material Table A; Fig. B).
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10 300 The VM denudation pattern, including the previously-reported lack of correlation with morphometric/climatic
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12 301 parameters, can be tentatively explained by two main features peculiar to this low-elevation mid-latitude mountain
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14 302 range. First, deciduous, resinous and mixed forests currently cover ~70% of its surface (Rochel, 2004), mostly on
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16 303 hillslopes and interfluvial areas (Fig. 2B). They were probably even more widespread before the Middle Age as
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18 304 forest clearance linked to past agricultural practices occurred at that time (Keller et al., 2023). Whilst vegetation
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20 305 has been increasingly considered as a controlling or modulating factor on CWD rates (e.g. Mandal et al., 2015;
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22 306 Olen et al., 2016; Schaller et al., 2018), Acosta et al. (2015) reported lower denudation rates in areas covered by
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24 307 dense vegetation. We likewise suggest that, in addition to this possible denudation lowering effect from dense
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26 308 vegetation, the VM's dense forest cover could have smoothed the variability in topographic surface erodibility.
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28 309 This would have resulted in lower amplitude of CWD rates that do not much differ across the thirteen investigated
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30 310 catchments, despite the contrasted ranges in elevation, slope, k_{sn} and annual precipitation.
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34 312 Second, and more importantly, the lithological heterogeneity encountered within the thirteen *steady-state*
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36 313 catchments possibly overprints the other controlling factors (Morel et al., 2003; Campforts et al., 2020). In the
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38 314 following we compare the crystalline- (#4, 5, 7, 8 and 20) and sedimentary-dominated (# 1, 9, 10, 15, 16 and 21)
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40 315 catchments (Figs. 3 and 6). Although mean k_{sn} value of the crystalline group ($\sim 58 \text{ m}^{0.9}$) is about three times higher
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42 316 than that of the sedimentary group ($\sim 19 \text{ m}^{0.9}$), ^{10}Be - and ^{26}Al -derived CWD do not significantly differ between
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44 317 both groups: $\sim 53/\sim 60 \text{ mm/ka}$ versus $\sim 51/\sim 52 \text{ mm/ka}$ in crystalline and sedimentary catchments, respectively
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46 318 (Supplementary Material, Fig. B). Despite an observed lower magnitude in relief incision (i.e. $\sim 150 \text{ m}$) in the
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48 319 sedimentary-dominated landscapes of the northern VM, denudation there is assumed to be as high as in the much
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50 320 more incised southern part (Figs. 1C and 2). The observed similarity in CWD rates obtained for both lithological
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52 321 groups thus implies that the erodibility of the homogeneous cover of Triassic sandstones should be much higher
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54 322 than that of Palaeozoic granites and gneisses. Note that all these catchments are located at the margins of the
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56 323 former glacier extents and, accordingly, must have experienced periglacial conditions during Quaternary cold
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3 324 stages. In addition, sandstone's enhanced erodibility due to periglacial processes has been already recognised in
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5 325 Palaeozoic, European low-mountain ranges (e.g. Ardennes in Belgium, Rixhon and Juvigné, 2010) and was
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7 326 recently quantified by cosmogenic nuclides in sandstone-dominated catchments for the central Appalachians (Del
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9 327 Vecchio et al., 2022).

10 11 12 328 13 14 329 **5.2. Understanding the impacts of former glaciations on cosmogenic nuclide disequilibrium**

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16 330 One of the main outcomes of this study is the spatial correlation between catchments in cosmogenic disequilibrium
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18 331 and the reconstructed former glaciations covering the southern VM. Our aim here is to assess the impact of
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20 332 glaciations on the depletion of the $^{26}\text{Al}/^{10}\text{Be}$ ratio through two processes, glacial shielding and reworking of
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22 333 previously buried sediment. Among all processes listed by Wittmann et al. (2020) resulting in depleted Al/Be ratio
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24 334 in river sediments, these two are the only ones that are relevant in the VM.

25 26 27 335 28 29 336 **5.2.1 $^{26}\text{Al}/^{10}\text{Be}$ ratio's depletion correlates with repeated glacial shielding**

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32 337 Although glacial shielding is one of the best documented processes to cause $^{26}\text{Al}/^{10}\text{Be}$ -ratio lowering in bedrock
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34 338 of high-latitude, formerly glaciated regions (e.g. Corbett et al., 2013; Strunk et al., 2017; Andersen et al., 2018;
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36 339 2020), it was barely considered when assessing CWD rates from modern stream sediments. In the following, we
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38 340 show a quantitative comparison between $^{26}\text{Al}/^{10}\text{Be}$ ratios in modern river sediments and former glacial covers in
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40 341 each catchment attributed to the LGM/MIS2 (Fig. 8A) and the penultimate glaciation/MIS6 (Fig. 8B) because
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42 342 their reconstruction is based on reliable and widespread field evidence (Flageollet, 2002).

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47 344 Main outputs of this comparison are twofold. First, significant correlations ($p\text{-values} \leq 0.0001$) are observed
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49 345 between $^{26}\text{Al}/^{10}\text{Be}$ ratio and the extent of past glacial cover during MIS2 ($R^2=0.53$) and 6 ($R^2=0.62$; Fig. 8A-B).
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51 346 All unglaciated catchments (i.e. $<10\%$ of ice cover during MIS2) cluster on the 6.7 production-ratio line, regardless
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53 347 the considered glacial period. The eleven formerly ice-covered catchments show an inverse general trend for both
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55 348 cold periods: the larger the former glacial cover in each catchment, the lower the $^{26}\text{Al}/^{10}\text{Be}$ ratio measured in its
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57 349 stream sediments (Fig. 8A-B). This observation underlines the probable link between long-lasting (i.e. 10^{3-4} years)
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59 350 and repetitive ice shielding and unsteadiness in the stream cosmogenic nuclide signal. One must bear in mind here

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3 351 that pre-MIS6 glaciations were documented in the VM's southern part (Flageollet, 2002; Supplementary Material,
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5 352 Fig. A2) and suggested at its southern margin (Ménillet, 2019). We thus propose that shielding in this area probably
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7 353 occurred during many cold periods of the Quaternary even if the correlated deposits of these pre-MIS6 glaciations
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9 354 can only be found in fluvial terrace sequences (e.g. Moselle and Ognon) located at the massif's outskirts (Harmand,
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12 355 2020). Second, the significance of these relationships increases with larger ice shielding between the two glacial
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14 356 periods: the ice-covered area roughly doubles between MIS2 (~730 km²) and 6 (~1570 km²; Fig. 1E). Note that
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16 357 three catchments were fully ice-covered during the LGM (i.e. >95%; Fig. 8B). We suggest here that this R² increase
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18 358 (0.53 to 0.62) is probably linked to distinct ice dynamics reflected in the spatial pattern peculiar to each cold stage
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21 359 (Flageollet, 2002; Fig. 1E). The LGM was characterised by a mosaic of valley glaciers with various ice-flow
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23 360 directions and lengths (up to ~40 km in the Moselle, #12), leaving large parts of the VM's main divide and
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25 361 secondary ridges ice-free (Supplementary Material, Fig. A2). By contrast, only a handful of small-dimensioned
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27 362 nunataks protruded the almost continuous MIS6's ice field characterised by a former SW-NE extent of ~60 km
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30 363 (Supplementary Material, Fig. A2).

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32 364 Although reconstructing the thermal nature of the former glacial masses in the southern VM is beyond the scope
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34 365 of this study, two lines of arguments based on previous studies using ¹⁰Be and ²⁶Al measurements could point to
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36 366 limited subglacial erosion and thus previous cold-based ice cover. First, if occurrence of cold-based glaciers is
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38 367 traditionally assigned to arctic regions (Copland, 2013), the highest ridges in the northern Appalachians, i.e. a
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41 368 similar setting to the southern VM, were covered by cold-based ice during the LGM (Bierman et al., 2015). Second
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43 369 and most important, depleted ²⁶Al/¹⁰Be ratios in bedrock and erratics from Greenland were well explained by
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45 370 prolonged and repeated burial beneath cold-based ice during glaciations (Corbett et al., 2013). The same holds for
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47 371 glacial tills shielded by cold-based ice on the Baffin Island, where the sampled grain size matches the sandy
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50 372 fraction studied here (Staiger et al., 2006). The depleted ²⁶Al/¹⁰Be ratios of these eight sandy samples (i.e. 1.3 to
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52 373 4.3) fall into a similar range to ours (1.4 to 5.2). Such depleted values would imply minimum burial times ranging
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54 374 from ~500 ka to ~3 Ma (Fig. 5; Table 1). If this continuous burial duration was indeed observed on Baffin Island,
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56 375 it is not so in the VM, whose relief has certainly been re-exposed to radiation during each interglacial period. It
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59 376 therefore seems highly probable that VM sediments were not correctly re-exposed to cosmic-rays between
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61 377 glaciations. Knudsen and Egholm's (2018) modelling results showed that very low ²⁶Al/¹⁰Be ratios may not be
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3 378 exclusively explained by prolonged burial under non-erosive ice, but may also reflect spatio-temporally varying
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5 379 bedrock exposure/burial and erosion histories. In addition to ice shielding, we therefore suggest that repeated,
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7 380 complex sediment routing systems during deglaciation and subsequent interglacial periods probably characterised
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9 381 the formerly glaciated catchments of the VM, thereby explaining the depleted $^{26}\text{Al}/^{10}\text{Be}$ ratios measured in the
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12 382 stream sediments.

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16 384 *5.2.2. $^{26}\text{Al}/^{10}\text{Be}$ ratio's depletion correlates with the amount of glacial sediment storage*

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18 385 Successive sediment burial and reworking within a sediment routing system, sometimes for long periods of time
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21 386 (i.e. $> 10^5$ years; Fülöp et al., 2020; Bhattacharjee et al., 2023) or rapid changes in denudation rates may yield to
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23 387 depleted $^{26}\text{Al}/^{10}\text{Be}$ ratio measured in the stream sediments. Although the cosmogenic nuclide signal is likely to be
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25 388 preserved from the source area to the sink in most cases (Wittmann et al., 2009), a handful of studies highlighted
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27 389 a significant decrease in $^{26}\text{Al}/^{10}\text{Be}$ ratios measured in stream sediments from glaciated catchments (Safran et al.,
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30 390 2005; Hippe et al., 2012; Grin et al., 2018; Wittmann et al., 2020; Zhang et al., 2021). Glacial environments are
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32 391 generally well-known to have complex sediment routing systems (e.g. Porter et al., 2019). Based on CWD rates
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34 392 inferred from both nuclides, Zhang et al. (2021) also identified this issue but did not provide further explanations
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36 393 regarding the correlated processes. This study presents, to our knowledge, the first quantitative comparison
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39 394 between $^{26}\text{Al}/^{10}\text{Be}$ ratios in modern river sediments and the surficial proportions of Quaternary deposits at the
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41 395 individual catchment scale (Fig. 8C-D).

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43 396 When all Quaternary sediments are considered, regardless of their nature, a significant correlation is observed
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45 397 ($R^2=0.33$; $p\text{-value}<0.01$; Fig. 8C). When only glacial/fluvio-glacial sediments are considered, the strongest
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47 398 correlation is observed among all our proxies to assess glacial impact on measured $^{26}\text{Al}/^{10}\text{Be}$ ratios ($R^2=0.67$; $p\text{-}$
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49 399 $\text{value}<0.0001$; Fig. 8D), including ice-cover shielding. Whereas all unglaciated catchments cluster on the 6.7
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52 400 production-ratio line, higher depletions of $^{26}\text{Al}/^{10}\text{Be}$ ratios are observed with increasing percentage of glacial
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54 401 sediments, peaking in catchment #2 (Fig. 8D). This suggests that (i) glacially-reworked deposits contain more
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56 402 $^{26}\text{Al}/^{10}\text{Be}$ -depleted material than fluvial and colluvial ones, and (ii) the greater the amount of glacial deposits
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59 403 present in the catchment, the greater the likelihood for the current river channel to rework these deposits.
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3 405 A closer look at the spatial distribution of glacial/fluvio-glacial deposits throughout the southern VM clearly
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5 406 highlights a W-E gradient, with the highest proportions of glacial/fluvio-glacial sediments encountered in the west-
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7 407 flowing catchments (i.e # 2, 12, 13, 14, 17, 19; Fig. 8D and Supplementary Material Fig. A1). One must recall
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9 408 here the geologically controlled W-E increasing gradient in mean catchment slope: 12.6 vs 16.9° to the west and
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11 409 the east of the main divide, respectively. Efficient sediment transfer in the east-flowing catchments can be assumed
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13 410 because of (i) shorter transport distances (Fig. 1B), (ii) higher mean slopes (Fig. 4B), (iii) much lower proportions
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15 411 of glacial deposits along their intra-massif reaches (Supplementary Material Fig. A1) and (iv) km-large, thick,
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17 412 Quaternary alluvial fans extending at the eastern Vosgian footslopes (e.g. catchments #1, 4, 6, 8, 18; Wuscher,
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19 413 2021). As a result, only one of the east-flowing catchments (#3) plots apart from the 6.7 line (Fig. 8).
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25 415 By contrast, among the six west-flowing, formerly glaciated catchments, five of them show depleted $^{26}\text{Al}/^{10}\text{Be}$
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27 416 ratios (Fig. 8). This probably reveals more complex sediment routing systems and the catchment #2 (i.e. the so-
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29 417 called Cleurie) with the most depleted ratio of our whole dataset (~1.4) epitomises this. First, if this catchment
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31 418 belongs to the group of catchments that were significantly and repeatedly covered by glaciers (#2, 12, 13, 14; Fig.
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33 419 8A-B), it clearly stands out because of the highest relative proportion of glacial/fluvio-glacial sediments (i.e. >40%
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35 420 vs $\leq 20\%$ for the rest; Fig. 8D). Though being the second smallest catchment investigated in this study (<80 km²;
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37 421 Table 1), the Cleurie accordingly hosts the highest density and largest diversity of glacial and glacial-related
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39 422 landforms, including e.g. end-moraines, kame terraces, lacustrine and deltaic deposits (Seret, 1967; Flageollet,
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41 423 2002). Second, thicknesses of valley-floor sediments exceeding 50 m, though undated, were revealed by drillings
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43 424 performed in the uppermost part of the catchment (BRGM). Third, petrographical and mineralogical analyses
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45 425 performed in some of these deposits reveal a partially allochthonous origin for some of the sampled materials, i.e.
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47 426 from the catchment located to the south (#13; Flageollet, 1971). Fourth, the peculiar spatial distribution and
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49 427 geometry of the best-preserved, i.e. tens of meters high, end moraines strongly point to a triple convergence of
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51 428 valley glaciers supposedly occurring during the LGM and peaking in the formation of a palaeolake in the
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53 429 catchment's upstream part (Flageollet, 2002). Fifth, a major flood occurred in the valley in 1770, whose damages
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55 430 were mapped in an archive document (Salmon, 1770). The latter reveals erosion and reworking of glacial material
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57 431 buried at the base of the highest moraines. Altogether, the concentration and diversity of glacial(-related)
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332 landforms along with the long-lasting storage of reworked and partially allochthonous material (i.e. “stop-and-
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333 go”, possibly similar to foreland glacial systems, e.g. Cogež et al., 2018) suggests a very complex sediment routing
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334 system in the Cleurie which yields the most depleted $^{26}\text{Al}/^{10}\text{Be}$ ratio of our dataset. More generally, our findings
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335 suggest that the sediments sampled in the SW catchments of the VM could have survived several glacial cycles.
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336 They might have been shielded by cold-based ice during glaciations, without being sufficiently re-exposed to
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337 cosmic rays during interglacial periods as they were buried underneath thick glacial sediments. Measuring low
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338 $^{26}\text{Al}/^{10}\text{Be}$ ratios in modern stream sediment also implies that the river network is currently partly supplied by
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339 depleted $^{26}\text{Al}/^{10}\text{Be}$ sedimentary material.
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23 5.3. Regional denudation in the Vosges and Black Forest Mountains

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25 342 At the regional scale, we compare our CWD rates with those previously obtained in SW Germany in the
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343 neighbouring Black Forest Mountains which shows a similar overall N-S structuration in lithology, elevation and
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344 morphometry. We recalculated fifty ^{10}Be -inferred CWD rates published by Morel et al. (2003), Meyer et al. (2010)
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345 and Glotzbach et al. (2013) using the methodology reported above to allow quantitative comparisons (see
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346 Supplementary Material, part 1.3). All sampled catchments are located within a similar latitude range than the VM
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347 (Fig. 9); they can thus be compared in terms of lithology, elevation and morphometry. The median (and standard
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348 deviation) ^{10}Be -derived CWD rates for the Black Forest Mountains and VM, regardless of the cosmogenic nuclide
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349 steadiness of the samples, appear at first glance remarkably similar: 52.6 ± 23.6 and 52.9 ± 14.4 mm/ka, respectively
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350 (Fig. 9). However, two complications considerably hamper a meaningful comparison between the two datasets.
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351 First, the average drainage area of the Black Forest Mountains catchments (~ 27 km²) is one order of magnitude
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352 smaller than for the VM catchments (~ 262 km²; Fig. 9A). Moreover, a majority of these small catchments are
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353 located in the headwaters of three main rivers draining the Black Forest Mountains, i.e. the Danube, Kinzig and
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354 Wutach. This strongly contrasts with our sampling strategy consisting in sampling all main rivers draining the
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355 VM. Second and most importantly, almost all catchments sampled by Morel et al. (2003) and Glotzbach et al.
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356 (2013) are located within the former MIS6 glacier extent (specific sampling approach to investigate the impact of
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357 glacial erosion and ice cover on CWD rates). However, the sole ^{10}Be measurement in modern stream sediments in
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358 these studies leave this twofold fundamental question open: considering a cosmogenic nuclide non-steady state
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359 (Glotzbach et al., 2013), how much disequilibrium characterises the sampled material there?
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9 361 **6. Conclusions and perspectives**

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12 362 From the massif-scale characterisation coupled to the paired-nuclide analysis to infer ^{10}Be - and ^{26}Al -based
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14 363 catchment-wide denudation rates across the Vosges Massif, we can conclude that:

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16 364 (i) elevation, slope, k_{sn} and precipitation are primarily organised along a N-S increasing trend while a subordinated
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18 365 E-W gradient occurs;

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21 366 (ii) ^{10}Be - and ^{26}Al -derived catchment-wide denudation rates range from 34 ± 1 to 66 ± 2 , and 41 ± 3 to 73 ± 7 mm/ka,
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23 367 respectively, in thirteen steady-state catchments that are in cosmogenic equilibrium;

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25 368 (iii) nine samples have depleted $^{26}\text{Al}/^{10}\text{Be}$ ratios (up to 1.4) and they mostly cluster in the Vosges Massif's area
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27 369 (SW) which was the most intensively glaciated during Quaternary cold stages. They reveal cosmogenic nuclide
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29 370 unsteadiness in modern river sediments and must be accordingly discarded when it comes to infer catchment-wide
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31 371 denudation rates, thereby precluding any conclusion about controlling factors at the whole massif scale;

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34 372 (iv) no relationship is reported between elevation, slope, k_{sn} and precipitation, and catchment-wide denudation
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36 373 rates in the steady-state catchments. Instead, lithological contrasts may control the pattern of denudation with a
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38 374 higher erodibility, especially under periglacial conditions, of the sandstone-dominated catchment compared to the
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40 375 crystalline-dominated catchments. We also suggest that the dense and almost ubiquitous vegetation cover reduces
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42 376 the variability in erodibility;

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45 377 (v) a statistically significant relationship is observed between the degree of depletion of the $^{26}\text{Al}/^{10}\text{Be}$ ratios and
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47 378 the spatial pattern of glaciation: the larger the former glacial cover in each catchment, the lower the $^{26}\text{Al}/^{10}\text{Be}$ ratio
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49 379 measured in its stream sediments. It underlines the link between long-lasting and repetitive ice shielding and
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51 380 unsteadiness in the stream cosmogenic signal. It also possibly suggests the past existence of cold-based glaciers in
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53 381 the Vosges Massif.

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56 382 (vi) an even more significant relationship is reported between the degree of depletion of the $^{26}\text{Al}/^{10}\text{Be}$ ratios and
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58 383 the increasing proportion of glacial and fluvio-glacial deposits within each catchment. It underlines the link
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60 384 between complex sediment routing systems in glacial environments and unsteadiness in the stream cosmogenic
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3 485 signal. This also well aligns with an E-W gradient: efficient sediment transfer versus longer sediment storage in
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5 486 the east- and west-flowing catchments, respectively. The catchment with the lowest $^{26}\text{Al}/^{10}\text{Be}$ ratio (1.4)
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7 487 remarkably matches the most complex sediment routing system from all sampled catchments.
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12 489 This study raises several key points. First, we argue for systematic measurements of ^{26}Al to complement ^{10}Be and
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14 490 to test the steady-state assumption when it comes to infer catchment-wide denudation rates from modern stream
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16 491 sediments, especially in formerly glaciated landscapes. Second, we show that 86% of the catchments containing
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18 492 more than 10% of glacial/fluvio-glacial deposits (6/7) have depleted $^{26}\text{Al}/^{10}\text{Be}$ ratio and are thus in cosmogenic
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20 493 nuclide disequilibrium. A good knowledge of the regional glacial history, including quantitative and chronological
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22 494 data, is thus prerequisite and this 10% occurrence of glacial/fluvio-glacial deposits could be used, at first glance,
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24 495 as a threshold value. Third, despite strong quantitative evidence of $^{26}\text{Al}/^{10}\text{Be}$ ratio depletion trough glacial related
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26 496 processes, further work is required to improve our interpretations. As for the impact of former ice covers, one
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28 497 should refine their spatial extents, assess their palaeothicknesses and chronologies. Abundant erratics and moraine
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30 498 deposits are key landforms here. ^{10}Be and ^{26}Al measurements of glacially polished bedrock should also be
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32 499 performed to assess glacial erosion efficiency and test the assumption of former cold-based glaciers. As for the
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34 500 impact of complex sediment-routing system, along stream ^{10}Be and ^{26}Al measurements should be undertaken in
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36 501 both east- and west flowing catchments. Dating valley-floor sediments and sediment budgeting of Vosgian valleys
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38 502 would bring key elements as well.
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45 504 **CRedit authorship contribution statement**

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47 505 **Timothée Jautzy:** Conceptualisation, Methodology, Investigation, Visualisation, Software, Writing – Original
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49 506 draft. **Gilles Rixhon:** Funding acquisition, Conceptualisation, Methodology, Investigation, Visualisation, Writing
50
51 507 – Original draft. **Régis Braucher:** Conceptualisation, Methodology, Investigation, Writing – Review & Editing.
52
53 508 **Romain Delunel:** Conceptualisation, Methodology, Writing – Review & Editing. **Pierre G. Valla:**
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55 509 Conceptualisation, Methodology, Writing – Review & Editing. **Laurent Schmitt:** Funding acquisition. **ASTER**
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57 510 **Team:** Formal analysis – AMS measurements.
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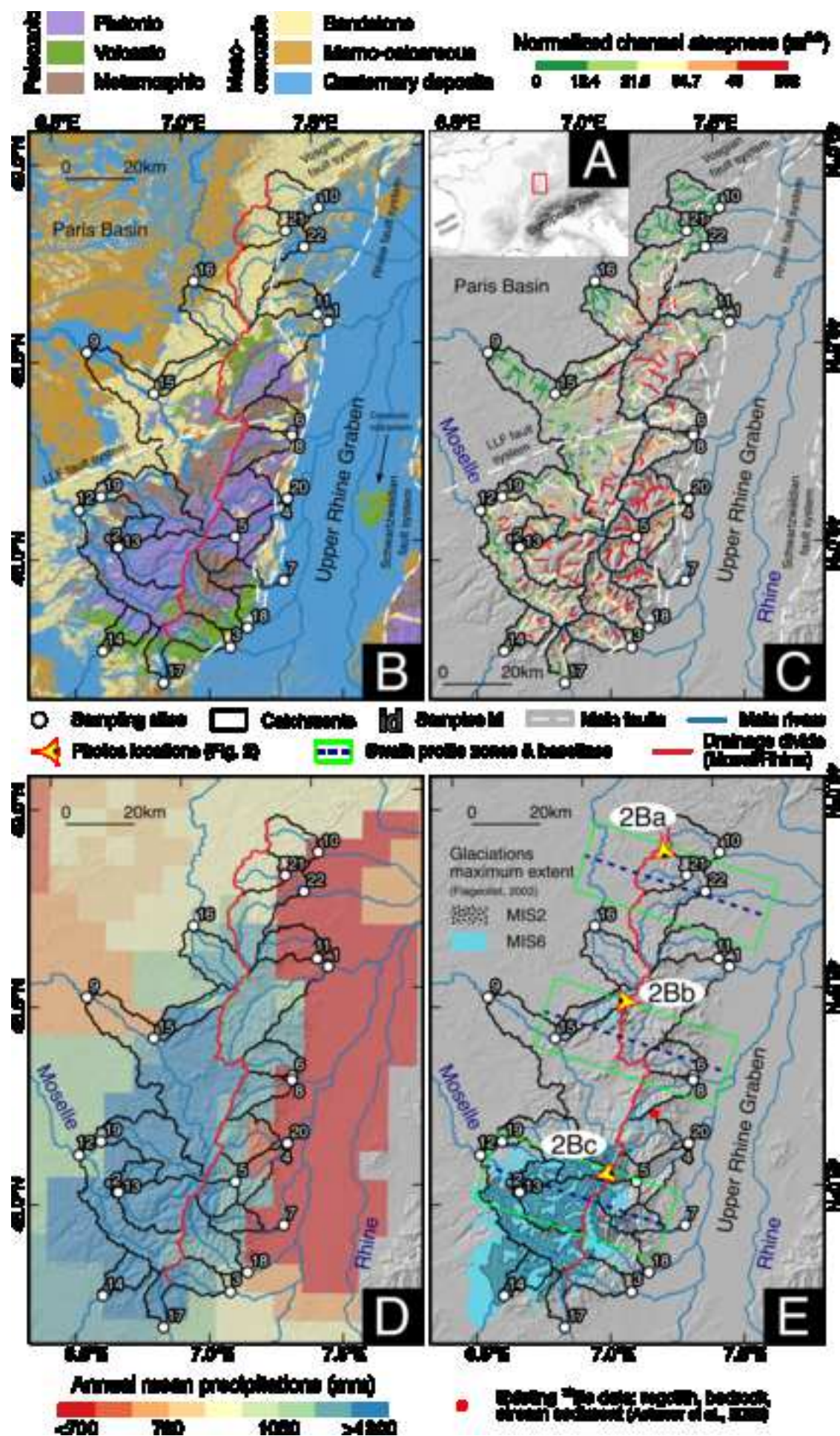
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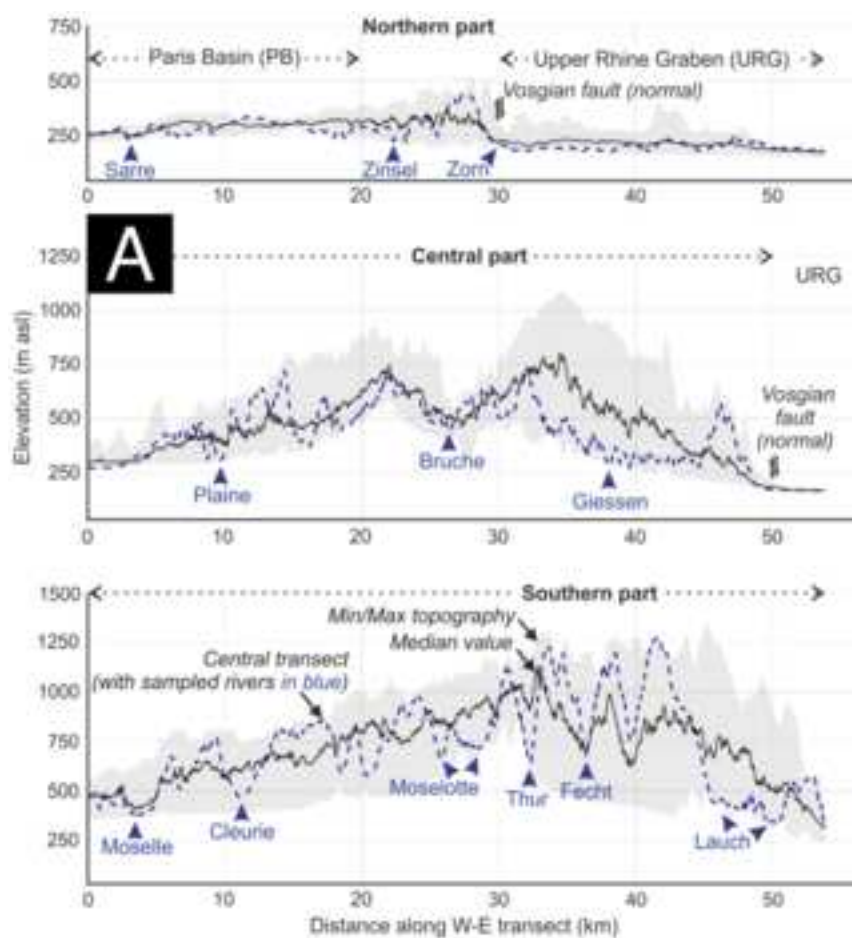
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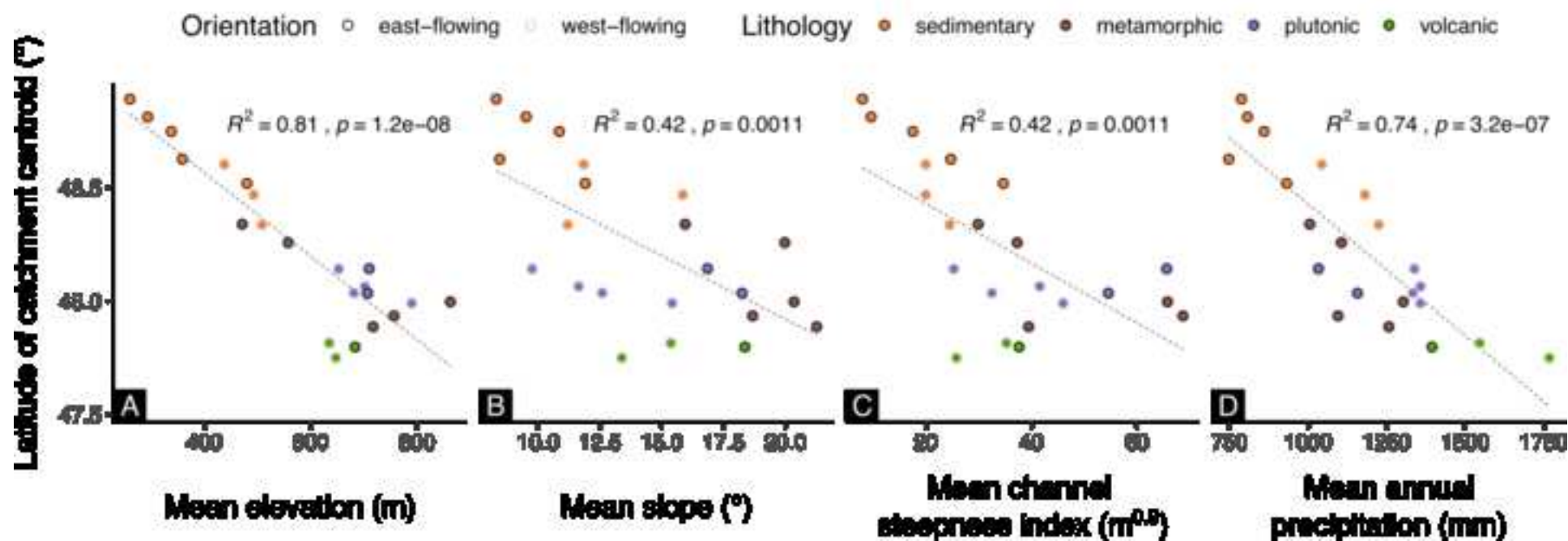
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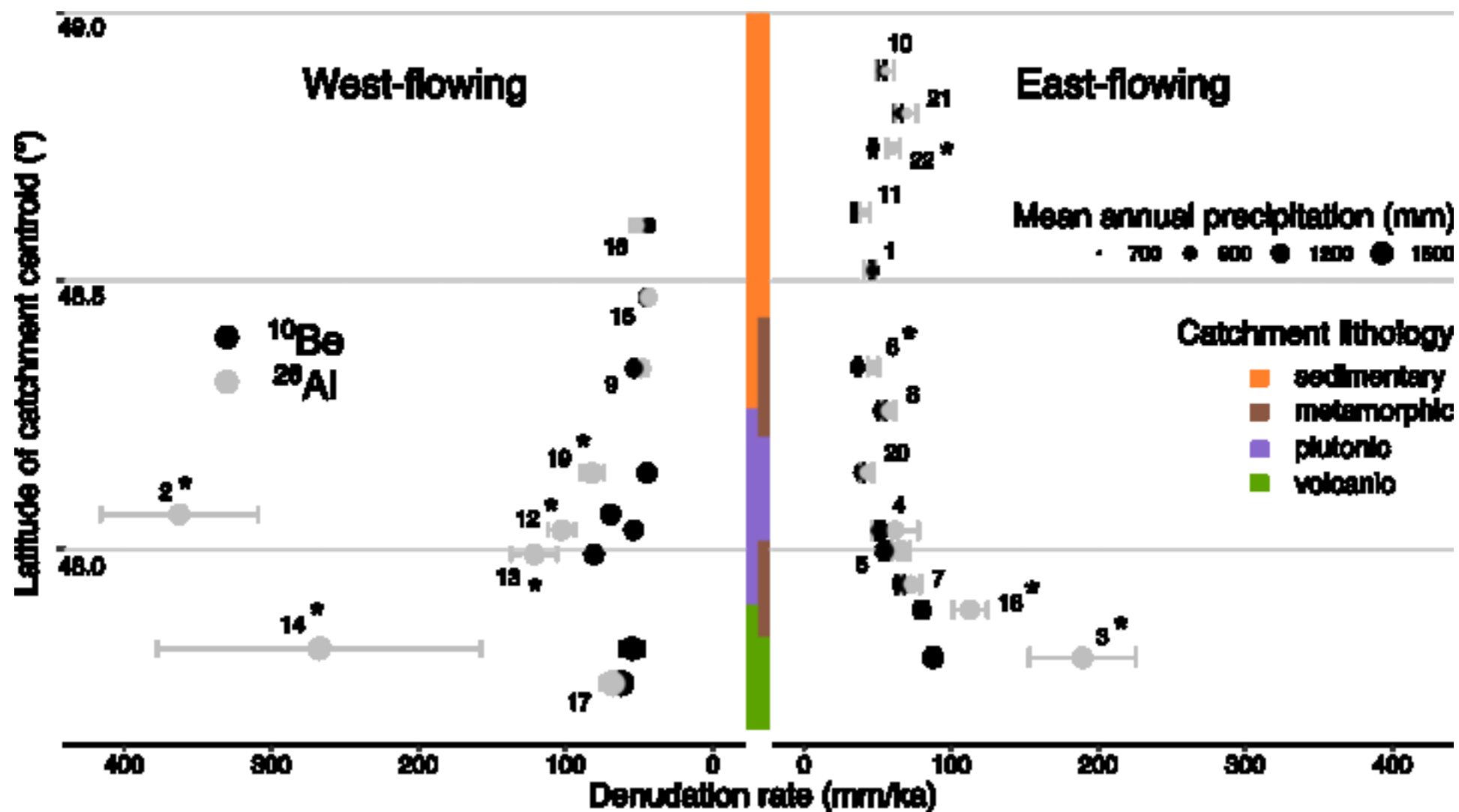
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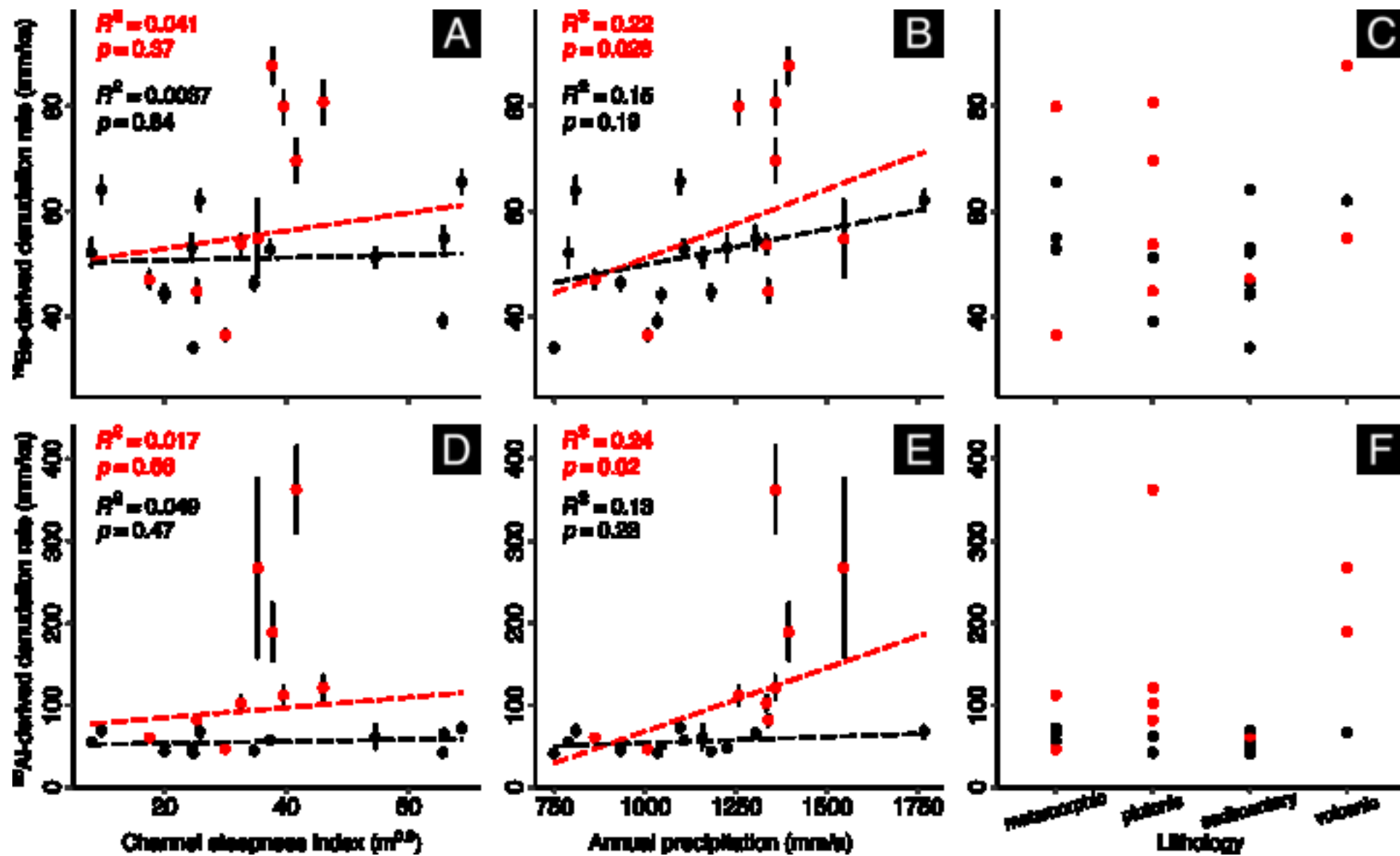
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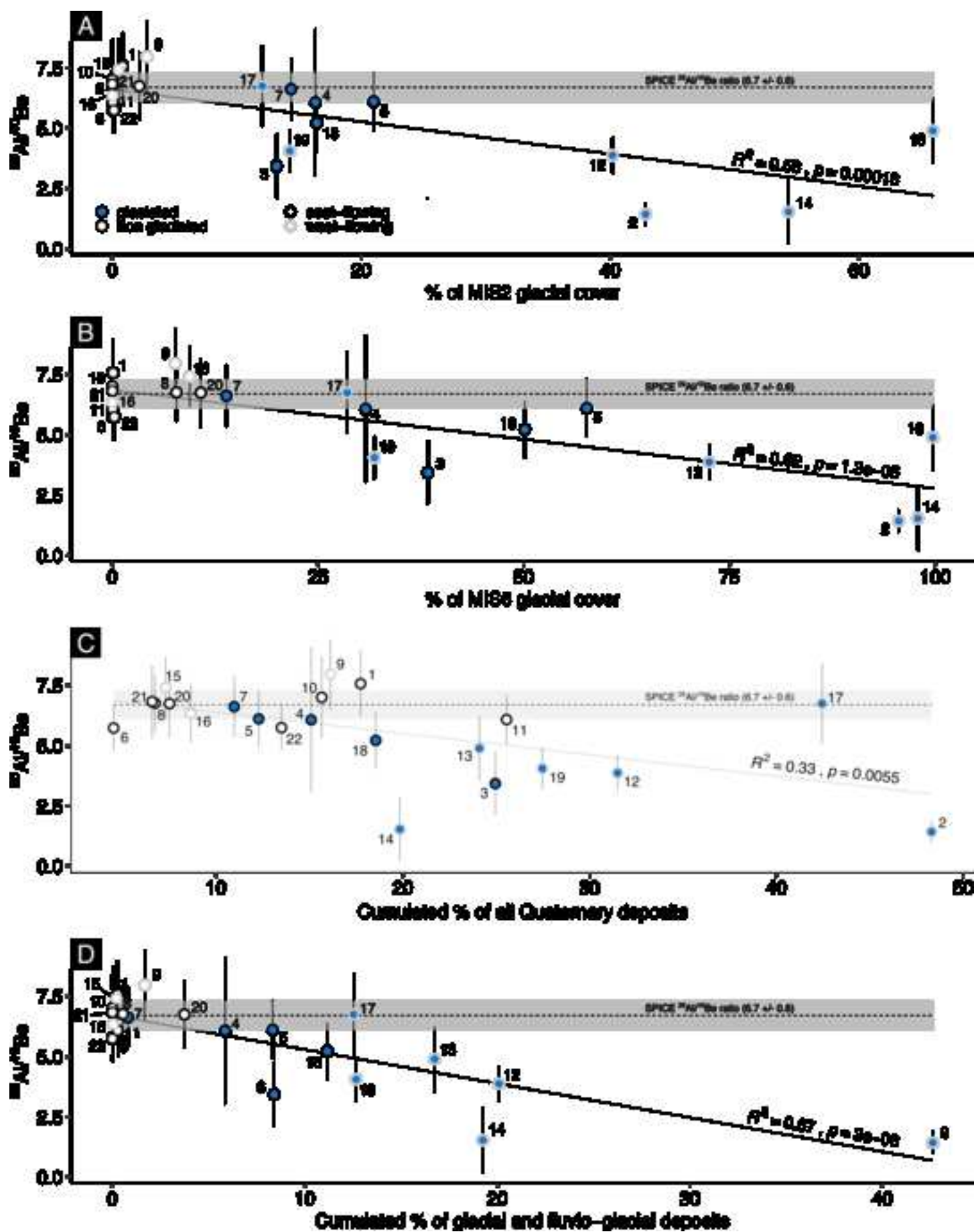












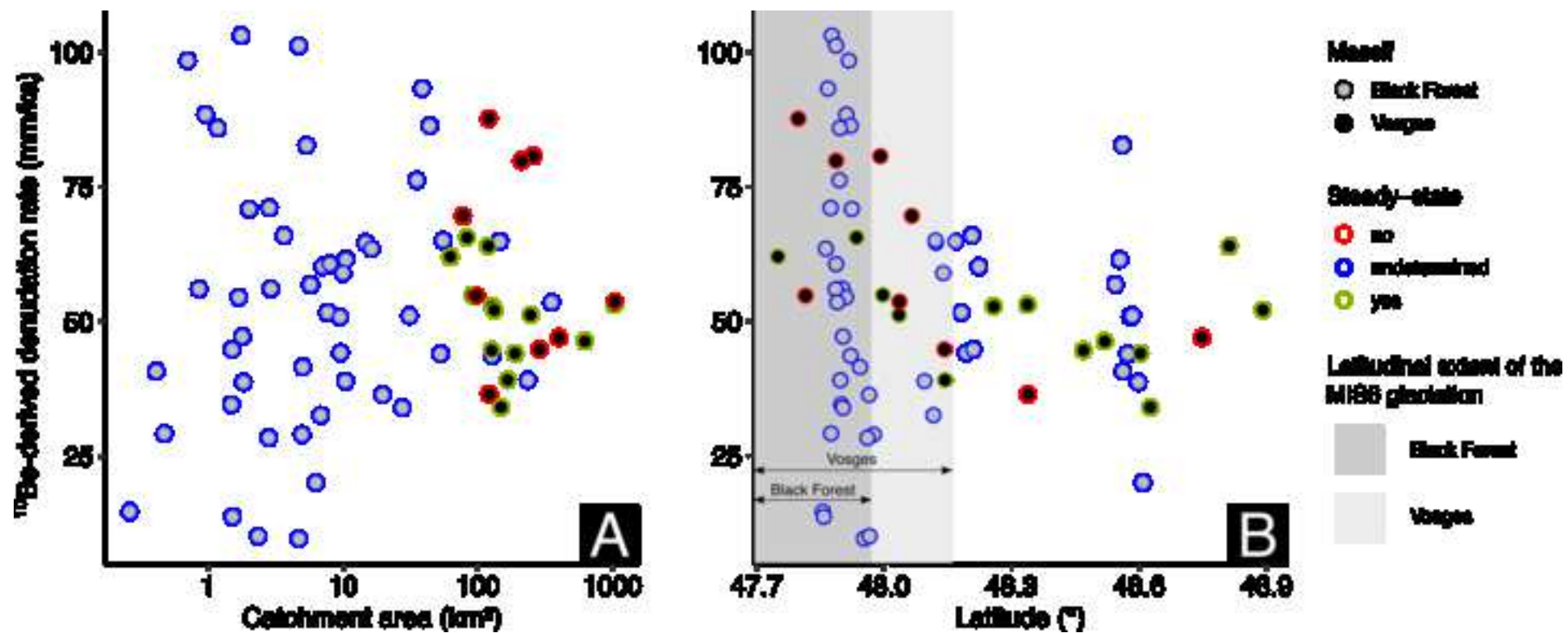


Table 1: Sample location, catchment characteristics, and cosmogenic nuclide results based on ^{10}Be and ^{26}Al measurements in quartz from stream sediment. ^aCatchment area associated with sample location, determined from a 25-m resolution DEM (BD ALTI). ^bMean elevation, ^cslope and ^dnormalised channel steepness index (using the Mudd et al.'s (2018) LSD Topotools software with a reference concavity of 0.45) are all computed from the same DEM. ^eMean annual precipitation determined from the national hydroclimatic database of Météo-France (Vidal et al., 2010) over the 1958-2009 period. ^fGlacial imprint based on the percentage of the catchment being ice-covered during MIS2 according to Flageollet (2002). >10% : yes; <10% : no. ^gMain lithology based on the hierarchical clustering analysis performed on bedrock lithologies and Quaternary deposits according to the same geological map (see Section 3.1). s: sedimentary, m: metamorphic, p: plutonic, v: volcanic. ^{h-i}Cosmogenic nuclide results for ^{10}Be and ^{26}Al .

Concentrations are corrected for a blank contribution. Analytical uncertainties (1σ) include both the AMS counting statistics and internal error (0.5% for ^{10}Be ; Arnold et al., 2010), chemical blank measurement and ^{27}Al measurements. Denudation rates are inferred using CosmoCalc version 3.1 (Vermeesch, 2007), based on the time-independent Lal (1991)/Stone (2000) scaling model. We used a density of 2.7g/cm^3 and did not consider topographic shielding. Mean production rates were calculated from the latitude of the catchments centroids and their mean elevation^b. Muonic productions were computed following Braucher et al. (2011). Reported uncertainties correspond to the 1σ analytical errors. Integration time corresponds to the time spent in the equivalent mean attenuation length (~60 cm).

^jMinimum burial time assuming a simple burial scenario for *non-steady-state* samples, calculated using CosmoCalc version 3.1 (Vermeesch, 2007).

Figure 1: Main characteristics of the Vosges Mountains (VM) with sample locations for cosmogenic nuclide analysis and their associated catchments. A: Location of the VM in Central Europe, forming the southwestern uplifted flank of the Upper Rhine Graben. B: Simplified geological map (1/50.000) according to the French Geological Survey (BRGM). C: Normalized channel steepness index of the hydrological network calculated in the sampled catchments using LSD Topotools software (Mudd et al., 2018) with a reference concavity of 0.45. D: Annual mean precipitation from the national hydroclimatic database of Météo-France (Vidal et al., 2010) over the 1958-2009 period. E: Glaciation maximum extents of the supposedly Marine Isotope Stage (MIS)2 and MIS6 cold periods (Flageollet, 2002). Green rectangles and yellow eyes match the swath profiles and the field pictures of Fig. 2, respectively. The red point pinpoints already existing catchment-wide denudation rates in the VM (Ackerer et al., 2016, 2022).

Figure 2: (A) North-South topographic gradient of the VM from swath profiles and (B) associated landscapes (locations in Figure 1E).

Figure 3: Lithological surficial proportions of each sampled catchments according to the 1/50.000 BRGM geological map. Catchments are classified according to their centroid's latitude. Letters in brackets corresponds to the results of the hierarchical clustering analysis performed on the bedrock lithologies and Quaternary deposits (see Section 3.1): sedimentary (s), metamorphic (m), plutonic (p) and volcanic (v).

Figure 4: Significant relationships between the latitude of catchment centroids and the morphometric/climatic factors in the VM. A: Mean elevation. B: Mean slope. C: Mean normalized channel steepness index. D: Mean annual precipitation. A second spatial gradient is also observed between the eastern-

and the western-flowing catchments. Note that statistics (R^2 ; p-value) are calculated using total least square regressions since the latitude does not depend on the controlling factors.

Figure 5: A: Two-nuclide diagram plotting $^{26}\text{Al}/^{10}\text{Be}$ ratios versus ^{10}Be concentrations. Sample ellipses correspond to 2σ analytical errors. Thirteen samples plot within the steady-erosion island and thus experienced steady-state erosion (grey ellipses). Nine samples plot below the steady-erosion island and thus experienced complex exposure histories (red ellipses). This plot was produced using CosmoCalc version 3.1 (Vermeesch, 2007), with the time-independent Lal (1991)/Stone (2000) scaling model. Muonic production was based on Braucher et al. (2011). B: Distribution of ^{26}Al -derived versus ^{10}Be -derived CWD rates. Error bars correspond to the 1σ analytical errors. Red and black points correspond to the *non-steady-state* and *steady-state catchments*, respectively (panel A). Note the logarithmic scales for both plots.

Figure 6: North-South distribution of CWD rates inferred from ^{10}Be (black points) and ^{26}Al (grey points) concentrations, along with the main lithologies and mean annual precipitation of the studied catchments. Horizontal error bars represent the 1σ uncertainty associated with the inferred CWD rates. *Non-steady-state catchments* are marked with an asterisk. The seven *non-steady-state samples* delivering the largest discrepancies in denudation values are all located in the southern part of the VM, regardless the catchment's lithology.

Figure 7: Relationships between CWD rates inferred from ^{10}Be (A-C) or ^{26}Al (D-F) concentrations and potential controlling factors: normalised channel steepness index, annual precipitation and main lithologies. (Red) black points correspond to the (*non*)-*steady-state samples*. Statistics are computed from linear regressions using the whole dataset (red lines) or the *steady-state* subset (black lines). A weak but significant relationship is observed between CWD rates and mean annual precipitation (p-value < 0.03). The significance of the relationship is lost when excluding the *non-steady-state samples* from the dataset (black lines).

Figure 8: Relationships between $^{26}\text{Al}/^{10}\text{Be}$ ratios and A: percentage of the catchment being ice-covered during MIS2, B: same for MIS6, C: cumulated percentage of all Quaternary deposits (i.e. alluvial, colluvial, glacial and fluvio-glacial deposits) and D: cumulated percentage of glacial and fluvio-glacial deposits. Glacial covers (A and B) are extracted from Flageollet's (2002) mapping. Quaternary sediment covers (C and D) are extracted from the BRGM's geological map. The vertical error bars correspond to the $^{26}\text{Al}/^{10}\text{Be}$ ratios at 1σ uncertainty. Blue points highlight the glaciated catchments (i.e. >10% of ice cover during MIS2). The horizontal dashed line corresponds to the surface production ratio recently determined by the SPICE project (6.7 ± 0.6 ; Fenton et al., 2022). We did not use the surface production ratio from Cosmocalc (Vermeesch, 2007) as it does not provide any uncertainty. The strongest significant linear regression ($R^2 = 0.67$, p-value = $3e-06$) is observed with the cumulated percentage of glacial and fluvio-glacial deposits (C).

Figure 9: Recalculated ^{10}Be -derived CWD rates in the Black Forest, compared to the ^{10}Be -derived CWD rates in the Vosges Massif, vs. A: catchment area and B: latitude of the catchment's centroid. The dark and light gray zones correspond to the latitudinal MIS6 glaciation extent of the Black Forest and the Vosges Massif, according to Flageollet (2002) and Ehlers et al. (2011), respectively.

Identification		Sample location		plain_text_2 Morphometric, climatic and lithological characteristics					
River name	id	Latitude (°)	Longitude (°)	Area ^a (km ²)	Mean elevation ^b (m)	Mean slope ^c (°)	Mean k_{sn} ^d (m ^{0.9})	Mean annual precipitation ^e (mm)	Glacial imprint ^f
Bruche	1	48.57	7.53	622.0	480.0	11.9	34.7	931.7	no
Cleurie	2	48.02	6.68	77.5	701.5	11.6	41.6	1357.6	yes
Doller	3	47.76	7.09	120.8	683.3	18.4	37.7	1394.2	yes
Fecht	4	48.12	7.33	246.0	707.1	18.2	54.6	1157.3	yes
Fecht	5	48.04	7.13	91.4	862.4	20.4	65.8	1302.8	yes
Giessen	6	48.30	7.36	121.4	471.3	15.9	29.9	1005.4	no
Lauch	7	47.92	7.30	83.1	756.6	18.7	68.7	1095.2	yes
Liepvrette	8	48.28	7.36	127.8	557.6	20.0	37.3	1105.8	no
Meurthe	9	48.52	6.60	1035.0	508.4	11.2	24.5	1224.5	no
Moder	10	48.86	7.51	132.4	260.2	8.3	8.0	788.0	no
Mossig	11	48.59	7.48	148.8	358.8	8.4	24.7	747.8	no
Moselle	12	48.12	6.54	1037.0	681.0	12.6	32.5	1333.3	yes
Moselotte	13	48.02	6.69	258.4	789.7	15.4	46.0	1357.0	yes
Ognon	14	47.76	6.61	96.9	634.8	15.4	35.3	1545.6	yes
Plaine	15	48.41	6.85	126.8	492.5	15.8	20.0	1180.7	no
Sarre	16	48.69	7.02	188.8	437.4	11.8	20.0	1043.7	no
Savoireuse	17	47.68	6.83	62.4	647.2	13.4	25.8	1766.9	yes
Thur	18	47.80	7.16	210.6	717.1	21.3	39.5	1256.8	yes
Vologne	19	48.15	6.63	286.8	652.6	9.7	25.3	1338.1	yes
Weiss	20	48.13	7.33	166.1	709.7	16.9	65.6	1033.0	no
Zinsel du Sud	21	48.80	7.38	119.8	293.7	9.5	9.6	807.4	no
Zorn	22	48.76	7.45	400.5	337.9	10.8	17.5	860.1	no

Main lithology ^g	¹⁰ Be results ^h			²⁶ Al results ⁱ			²⁶ Al/ ¹⁰ Be
	[¹⁰ Be] 10 ⁴ (at/g) qtz	¹⁰ Be - denudation rate (mm/ka)	¹⁰ Be - integration time (ka)	[²⁶ Al] 10 ⁴ (at/g) qtz	²⁶ Al - denudation rate (mm/ka)	²⁶ Al - integration time (ka)	
s	9.8 +/- 0.3	46 +/- 1	13.0	74 +/- 6	45 +/- 4	13.4	7.6 +/- 0.7
p	7.7 +/- 0.5	70 +/- 4	8.6	11.1 +/- 2	363 +/- 53	1.7	1.4 +/- 0.2
v	6.1 +/- 0.2	88 +/- 4	6.8	20.8 +/- 4	189 +/- 36	3.2	3.4 +/- 0.7
p	10.5 +/- 0.4	51 +/- 2	11.7	63.8 +/- 16	62 +/- 16	9.7	6.1 +/- 1.5
m	11 +/- 0.5	55 +/- 2	10.9	67.3 +/- 6	66 +/- 6	9.1	6.1 +/- 0.6
m	12.2 +/- 0.4	36 +/- 1	16.5	70.2 +/- 5	47 +/- 4	12.8	5.7 +/- 0.5
m	8.5 +/- 0.3	66 +/- 2	9.1	56.5 +/- 5	73 +/- 7	8.3	6.6 +/- 0.6
m	9.1 +/- 0.3	53 +/- 2	11.4	61.6 +/- 5	57 +/- 5	10.5	6.8 +/- 0.6
s	8.7 +/- 0.4	53 +/- 3	11.3	69.5 +/- 5	49 +/- 4	12.3	8 +/- 0.7
s	7.3 +/- 0.4	52 +/- 3	11.5	51.5 +/- 6	55 +/- 6	10.9	7 +/- 0.9
s	12 +/- 0.4	34 +/- 1	17.6	73 +/- 6	41 +/- 3	14.5	6.1 +/- 0.5
p	9.8 +/- 0.3	54 +/- 2	11.2	38.1 +/- 3	103 +/- 9	5.8	3.9 +/- 0.4
p	7.1 +/- 0.4	81 +/- 4	7.4	34.9 +/- 4	121 +/- 16	4.9	4.9 +/- 0.7
v	9.3 +/- 1.3	55 +/- 8	11.0	14.2 +/- 6	267 +/- 110	2.2	1.5 +/- 0.7
s	10.2 +/- 0.3	45 +/- 2	13.4	75.9 +/- 6	44 +/- 4	13.6	7.4 +/- 0.6
s	9.9 +/- 0.3	44 +/- 1	13.6	63 +/- 6	51 +/- 5	11.7	6.3 +/- 0.6
v	8.3 +/- 0.3	62 +/- 2	9.7	55.9 +/- 7	67 +/- 8	8.9	6.8 +/- 0.8
m	6.8 +/- 0.3	80 +/- 3	7.5	35.6 +/- 4	113 +/- 12	5.3	5.2 +/- 0.6
p	11.5 +/- 0.6	45 +/- 2	13.4	46.6 +/- 4	82 +/- 8	7.3	4.1 +/- 0.4
p	13.7 +/- 0.5	39 +/- 1	15.3	92.5 +/- 9	42 +/- 4	14.2	6.7 +/- 0.7
s	6.2 +/- 0.2	64 +/- 3	9.4	42.1 +/- 4	69 +/- 7	8.7	6.8 +/- 0.8
s	8.6 +/- 0.3	47 +/- 2	12.8	49.7 +/- 4	61 +/- 4	9.9	5.7 +/- 0.5

Minimum burial time ^j (ka)
3351.3
1570.7
501.2
1310.3
832.5
3198.7
701.4
1210.6
508.7

Identification		Sample location		feuille 1 Morphometric, climatic and lithological characteristics					
River name	id	Latitude (°)	Longitude (°)	Area ^a (km ²)	Mean elevation ^b (m)	Mean slope ^c (°)	Mean k_{sn}^d (m ^{0.9})	Mean annual precipitations ^e (mm)	Glacial imprint ^f
Bruche	1	48.57	7.53	622.0	480.0	11.9	34.7	931.7	no
Cleurie	2	48.02	6.68	77.5	701.5	11.6	41.6	1357.6	yes
Doller	3	47.76	7.09	120.8	683.3	18.4	37.7	1394.2	yes
Fecht	4	48.12	7.33	246.0	707.1	18.2	54.6	1157.3	yes
Fecht	5	48.04	7.13	91.4	862.4	20.4	65.8	1302.8	yes
Giessen	6	48.30	7.36	121.4	471.3	15.9	29.9	1005.4	no
Lauch	7	47.92	7.30	83.1	756.6	18.7	68.7	1095.2	no
Liepvrette	8	48.28	7.36	127.8	557.6	20.0	37.3	1105.8	no
Meurthe	9	48.52	6.60	1035.0	508.4	11.2	24.5	1224.5	no
Moder	10	48.86	7.51	132.4	260.2	8.3	8.0	788.0	no
Mossig	11	48.59	7.48	148.8	358.8	8.4	24.7	747.8	no
Moselle	12	48.12	6.54	1037.0	681.0	12.6	32.5	1333.3	yes
Moselotte	13	48.02	6.69	258.4	789.7	15.4	46.0	1357.0	yes
Ognon	14	47.76	6.61	96.9	634.8	15.4	35.3	1545.6	yes
Plaine	15	48.41	6.85	126.8	492.5	15.8	20.0	1180.7	no
Sarre	16	48.69	7.02	188.8	437.4	11.8	20.0	1043.7	no
Savoureuse	17	47.68	6.83	62.4	647.2	13.4	25.8	1766.9	yes
Thur	18	47.80	7.16	210.6	717.1	21.3	39.5	1256.8	yes
Vologne	19	48.15	6.63	286.8	652.6	9.7	25.3	1338.1	yes
Weiss	20	48.13	7.33	166.1	709.7	16.9	65.6	1033.0	no
Zinsel du Sud	21	48.80	7.38	119.8	293.7	9.5	9.6	807.4	no
Zorn	22	48.76	7.45	400.5	337.9	10.8	17.5	860.1	no

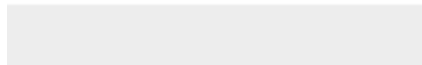
Main lithology ^g	¹⁰ Be results ^h			feuille 1 ²⁶ Al results ⁱ			²⁶ Al/ ¹⁰ Be
	[¹⁰ Be] 10 ⁴ (at/g) qtz	¹⁰ Be - denudation rate (mm/ka)	¹⁰ Be - integration time (ka)	[²⁶ Al] 10 ⁴ (at/g) qtz	²⁶ Al - denudation rate (mm/ka)	²⁶ Al - integration time (ka)	
s	9.8 +/- 0.3	46 +/- 1	13.0	74 +/- 6	45 +/- 4	13.4	7.6 +/- 0.7
p	7.7 +/- 0.5	70 +/- 4	8.6	11.1 +/- 2	363 +/- 53	1.7	1.4 +/- 0.2
v	6.1 +/- 0.2	88 +/- 4	6.8	20.8 +/- 4	189 +/- 36	3.2	3.4 +/- 0.7
p	10.5 +/- 0.4	51 +/- 2	11.7	63.8 +/- 16	62 +/- 16	9.7	6.1 +/- 1.5
m	11 +/- 0.5	55 +/- 2	10.9	67.3 +/- 6	66 +/- 6	9.1	6.1 +/- 0.6
m	12.2 +/- 0.4	36 +/- 1	16.5	70.2 +/- 5	47 +/- 4	12.8	5.7 +/- 0.5
m	8.5 +/- 0.3	66 +/- 2	9.1	56.5 +/- 5	73 +/- 7	8.3	6.6 +/- 0.6
m	9.1 +/- 0.3	53 +/- 2	11.4	61.6 +/- 5	57 +/- 5	10.5	6.8 +/- 0.6
s	8.7 +/- 0.4	53 +/- 3	11.3	69.5 +/- 5	49 +/- 4	12.3	8 +/- 0.7
s	7.3 +/- 0.4	52 +/- 3	11.5	51.5 +/- 6	55 +/- 6	10.9	7 +/- 0.9
s	12 +/- 0.4	34 +/- 1	17.6	73 +/- 6	41 +/- 3	14.5	6.1 +/- 0.5
p	9.8 +/- 0.3	54 +/- 2	11.2	38.1 +/- 3	103 +/- 9	5.8	3.9 +/- 0.4
p	7.1 +/- 0.4	81 +/- 4	7.4	34.9 +/- 4	121 +/- 16	4.9	4.9 +/- 0.7
v	9.3 +/- 1.3	55 +/- 8	11.0	14.2 +/- 6	267 +/- 110	2.2	1.5 +/- 0.7
s	10.2 +/- 0.3	45 +/- 2	13.4	75.9 +/- 6	44 +/- 4	13.6	7.4 +/- 0.6
s	9.9 +/- 0.3	44 +/- 1	13.6	63 +/- 6	51 +/- 5	11.7	6.3 +/- 0.6
v	8.3 +/- 0.3	62 +/- 2	9.7	55.9 +/- 7	67 +/- 8	8.9	6.8 +/- 0.8
m	6.8 +/- 0.3	80 +/- 3	7.5	35.6 +/- 4	113 +/- 12	5.3	5.2 +/- 0.6
p	11.5 +/- 0.6	45 +/- 2	13.4	46.6 +/- 4	82 +/- 8	7.3	4.1 +/- 0.4
p	13.7 +/- 0.5	39 +/- 1	15.3	92.5 +/- 9	42 +/- 4	14.2	6.7 +/- 0.7
s	6.2 +/- 0.2	64 +/- 3	9.4	42.1 +/- 4	69 +/- 7	8.7	6.8 +/- 0.8
s	8.6 +/- 0.3	47 +/- 2	12.8	49.7 +/- 4	61 +/- 4	9.9	5.7 +/- 0.5

Minimum burial time (ka)	Burial-corrected denudation rate (mm/ka)	Minimum burial time (ka)	Burial-corrected denudation rate (mm/ka)	error
3351.3	12 +/- 6	3351.3	12.4	6.0
1570.7	39 +/- 54	1570.7	39.4	53.7
501.2	28 +/- 25	501.2	28.1	25.0
1310.3	27 +/- 20	1310.3	27.4	19.9
832.5	53 +/- 69	832.5	52.9	69.3
3198.7	10 +/- 15	3198.7	10.4	14.7
701.4	56 +/- 64	701.4	55.9	64.1
1210.6	24 +/- 20	1210.6	24.0	19.6
508.7	36 +/- 31	508.7	36.2	31.2



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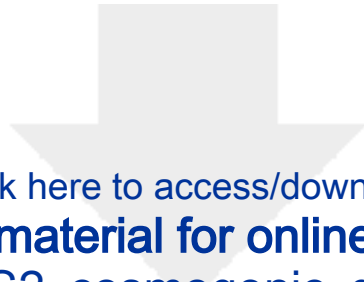




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Supplementary material for online publication only

Table_S2_cosmogenic_data.ods



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