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## **Regolith evolution on the millennial timescale from combined U-Th-Ra isotopes and in situ cosmogenic $^{10}\text{Be}$ analysis in a weathering profile (Strengbach catchment, France)**

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1 **Regolith evolution on the millennial timescale from combined U-Th-Ra isotopes and in situ**  
2 **cosmogenic <sup>10</sup>Be analysis in a weathering profile (Strengbach catchment, France)**

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21

22 **Abstract**

23 U-Th-Ra disequilibria, cosmogenic in situ  $^{10}\text{Be}$  concentrations and major and trace element  
24 concentrations have been analyzed in a 2 m-deep weathering profile sampled at the summit  
25 of the granitic Strengbach catchment (France). The data have been used to independently  
26 estimate both the long-term regolith production and denudation rates and the weathering  
27 and erosion rates. Modeling of the  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$ - $^{226}\text{Ra}$  disequilibrium variations in the  
28 lower part of the profile yields a regolith production rate of  $12\pm 4$  mm/kyr ( $30\pm 10$  T/km<sup>2</sup>/yr),  
29 while modeling of the high-resolution  $^{10}\text{Be}$  concentration profile leads to an exposure age of  
30  $19.7\pm 2.2$  kyr, an inherited concentration of  $15,000\pm 1,000$  at/g in quartz and a mean  
31 denudation rate of  $22\pm 10$  mm/kyr ( $37\pm 15$  T/km<sup>2</sup>/yr). The consistency between production  
32 and denudation rates suggests that, on a millennial timescale, the regolith mass balance at  
33 the summit of the catchment is close to a steady state, even if the watershed may have been  
34 impacted by Quaternary climatic changes and by recent anthropogenic perturbations (e.g.,  
35 20<sup>th</sup> century acid rain and recent afforestation efforts). The results also indicate that physical  
36 erosion is likely the dominant long-term process of regolith denudation in the catchment.  
37 Furthermore, the comparison of the long-term production and denudation rates and of  
38 weathering and erosion rates determined from the depth profile analyses with the current  
39 weathering and erosion rates estimated at the outlet of the watershed based on monitoring  
40 of the water chemistry and sediment fluxes suggests that physical erosion may have varied  
41 more than the chemical weathering flux during the last 150 kyr. Although very few other  
42 sites with U-series, in situ  $^{10}\text{Be}$  and stream monitoring data are available for comparison, the  
43 current data suggest that (1) the mass balance steady state of regolith might be commonly  
44 achieved in soil mantled landscapes and (2) physical erosion has varied much more than  
45 chemical weathering in mid-mountain catchments over the last 10-150 kyr. These results  
46 highlight the importance of the combined analysis of U-series nuclides and in situ  $^{10}\text{Be}$  in the  
47 same weathering profile for the determination of key geomorphic parameters, which are  
48 important to constraining landscape stability and the responses of landscapes to natural or  
49 anthropogenic forcing.

50 **Keywords**

51 Regolith, weathering, denudation, U-series nuclides, in situ  $^{10}\text{Be}$ .

52 **1- Introduction**

53 Regolith production and denudation rates, which correspond to the rate at which bedrock is  
54 weathered into mobile regolith and removed by chemical and physical processes, are key  
55 parameters in the evaluation of landscape stability and the responses of landscapes to  
56 natural or anthropogenic forcing (e.g., Brantley et al., 2007; Banwart et al., 2011). The  
57 analytical developments made over the last decades for precisely analyzing the U-series  
58 nuclides (i.e.,  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$ ) in geological and environmental samples have led to the  
59 development of the study of U-series nuclides in soils and weathering profiles and to the  
60 definition of a theoretical framework for quantifying regolith production rates from the  
61 variations in radioactive disequilibria along a weathering profile (Dequincey et al., 2002;  
62 Chabaux et al., 2003, 2013; Dosseto et al., 2008, 2012; Ma et al., 2010). These studies were  
63 mainly based on the analysis of  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$  nuclides and the use of the activity ratios  
64 ( $^{234}\text{U}/^{238}\text{U}$ )-( $^{230}\text{Th}/^{234}\text{U}$ ) (activity ratios will be noted hereafter with parentheses). More  
65 recently, some studies have also included the analysis of the  $^{226}\text{Ra}$  nuclide and hence the use  
66 of the ( $^{226}\text{Ra}/^{230}\text{Th}$ ) ratio (Chabaux et al., 2013; Gontier et al., 2015). Similarly, the in situ  $^{10}\text{Be}$   
67 depth profile methodology has enabled the estimation of both exposure age and mean  
68 denudation rate from cosmogenic isotope inventories (e.g., Brown et al., 1995; Braucher et  
69 al., 2009). This approach has been widely used to constrain the ages of alluvial terraces and  
70 fans (Anderson et al., 1996; Schaller et al., 2002; Brocard et al., 2003), as well as the long-  
71 term denudation rates of regolith (Small et al., 1999; Heimsath et al., 2000; Ferrier and  
72 Kirchner, 2008; Cui et al., 2016). However, very few studies have combined a detailed  
73 analysis of U-Th-Ra isotopes with cosmogenic in situ  $^{10}\text{Be}$  in a single weathering profile  
74 extending from the topsoil to the bedrock. It is the aim of this work to highlight the potential  
75 of combining these two approaches to independently constrain both production and

76 denudation rates of regolith and to show that information associated with geochemical  
77 mobility can be used to discuss the long-term evolution of the regolith.

## 78 **2- Site presentation and sampling strategy**

79 The study is performed on the Strengbach catchment, which constitutes the “Observatoire  
80 Hydrogéochimique de l’Environnement” (OHGE), one of the French critical zone  
81 observatories (<http://rnbv.ipgp.fr>). It is a small watershed of 0.8 km<sup>2</sup> located in the Vosges  
82 massif (northeastern France; Figure 1A). With altitudes ranging from 883 to 1147 m (Figures  
83 1B and 1C), the current climate is mountainous-oceanic, with a mean annual rainfall and  
84 temperature of 1400 mm and 6°C, respectively (Viville et al., 2012). The Vosges massif  
85 experienced Pleistocene glaciations in a similar way to other central European mountains,  
86 such as the Black Forest and the Bavarian Forest (Heyman et al., 2013). In accordance with  
87 regional climatic studies (e.g., Leroy et al., 2000), cold conditions probably persisted at the  
88 altitude of the Strengbach catchment well after the Late Glacial Maximum (LGM), and the  
89 Vosges forest cover likely developed only at the onset of the Holocene. The Strengbach site  
90 has been affected by anthropogenic deforestation associated with pastoralism, likely  
91 beginning in the Bronze Age and lasting until the end of the 19<sup>th</sup> century (Etienne et al.,  
92 2013). In the 20<sup>th</sup> century, the lower grazing pressure led to natural and artificial  
93 afforestation in several places in the Vosges massif. In the Strengbach catchment, the return  
94 to a densely forested cover is due to the planting of spruce stands at the beginning of the  
95 20<sup>th</sup> century. The catchment is currently covered with a mixed spruce and beech forest  
96 interspersed with small clearings. The bedrock is a base-poor late Hercynian granite and is  
97 covered by a 50 to 100 cm-thick brown acidic soil (Hyperdystric Cambisol ; WRB 2006). The  
98 granitic bedrock was hydrothermally overprinted, with the degree of hydrothermal

99 alteration decreasing from the northern to the southern part of the watershed (Fichter et al.,  
100 1998).

101 To investigate the recent Quaternary weathering of the granitic bedrock and the denudation  
102 rate of the soil, a sampling profile has been collected on the summit of the less  
103 hydrothermally altered part of the watershed. This location avoids the presence of colluvial  
104 deposition and corresponds to the best strategy ensuring that saprolite and soil are  
105 genetically linked to the underlying bedrock and likely formed along the main vertical  
106 weathering direction. A 2 m-deep and 3 m-wide pit was dug, and the samples were collected  
107 in the middle part of the pit front (Figure 1D). Thirty-two bulk samples of  $\approx 5$  kg each were  
108 collected along the profile with a sample every 5 cm within the soil and the saprolite and  
109 every 10 cm within the fractured bedrock. The large mass collected per sample is necessary  
110 to ensure a representative analysis of the weathering profile developed on the coarse-  
111 grained granite (Gy, 1992). For each sample in the upper part of the profile (0-100 cm), the  
112 fine fraction density was obtained by weighing a known volume of soil sampled with a steel  
113 cylinder. The proportion of blocks, defined here as coarse fragments ( $>10$  cm), was  
114 estimated via macroscopic description and photos. The bulk density of each sample was  
115 then calculated by combining the fine fraction density and the density of blocks with respect  
116 to the proportion of blocks in the horizon. For the blocks, a constant density of  $2700 \text{ kg/m}^3$  is  
117 used, on the basis of granitic bedrock density measurements. A sequential crushing and  
118 rigorous quartering process was then performed to obtain representative subsamples for  
119 mineralogical analyses by X-ray diffraction (XRD), and for geochemical and isotopic analyses.

## 120 **3- Analytical methods**

### 121 **3.1 Mineralogical and major and trace element analysis**

122 The determination of the sample mineralogical compositions are given in Electronic  
123 Appendix A. Six thin sections of rock samples were collected at regular intervals within the  
124 granitic bedrock (Figure 1D). Major and trace element concentrations of the samples were  
125 analyzed following the technique classically used at the LHyGeS (Strasbourg, France; e.g.,  
126 Prunier et al., 2015). The quality of the analysis was checked by measuring the San Joaquin  
127 soil standard (NIST SRM 2709a). The overall uncertainty is <5 % for the major element and  
128 <10 % for the trace element concentrations (Table 1).

### 129 **3.2 U-Th-Ra isotopes**

130 For the U-Th-Ra analysis, each subsample consists of approximately 100 mg of 53  $\mu\text{m}$   
131 powdered bulk sample. Subsamples were weighed and spiked with both a mixed artificial  
132  $^{233}\text{U}$ - $^{229}\text{Th}$  spike and a  $^{228}\text{Ra}$  enriched spike to measure the isotopic ratios and U-Th-Ra  
133 concentrations by isotopic dilution, using the protocol developed in the LHyGeS (Strasbourg,  
134 France; Granet et al., 2010; Chabaux et al., 2013; Pelt et al., 2013). The powders were first  
135 digested in PFA beakers using combinations of HF,  $\text{HNO}_3$ , HCl,  $\text{HClO}_4$  and  $\text{H}_3\text{BO}_3$  acids at 100-  
136 200°C, and U, Th, and Ra were then separated and purified by ion exchange chromatography  
137 using Biorad anionic resin. U and Th isotopes were measured on a plasma mass spectrometer  
138 (MC-ICP-MS Neptune), and the  $^{226}\text{Ra}$  isotope was measured on a thermal ionization mass  
139 spectrometer (TIMS Triton, Table 2). The precision and accuracy of the analyses are checked  
140 by regular analysis of liquid standards (HU1 U, IRMM036 Th and  $^{228}\text{Ra}$ -spike), the rock  
141 standard BCR-2 and sample replicates. The ( $^{234}\text{U}/^{238}\text{U}$ ) activity ratio was  $0.9992\pm 0.0018$  for  
142 HU1 ( $2\sigma$ , N=19) and the  $^{232}\text{Th}/^{230}\text{Th}$  isotope ratio was  $326284\pm 2855$  for IRMM036 ( $2\sigma$ , N=21).  
143 The ( $^{234}\text{U}/^{238}\text{U}$ ), ( $^{230}\text{Th}/^{234}\text{U}$ ), ( $^{230}\text{Th}/^{232}\text{Th}$ ) and ( $^{226}\text{Ra}/^{230}\text{Th}$ ) activity ratios for BCR-2 were  
144  $1.0028\pm 0.0018$ ,  $1.0045\pm 0.0104$ ,  $0.8824\pm 0.0076$  and  $0.9946\pm 0.0078$  ( $2\sigma$ , N=2), respectively.

145 The mean U and Th blank concentrations were 46 pg and 157 pg, respectively, whereas the  
146 Ra concentrations were below the detection limit. These blank concentrations were all  
147 negligible compared to the measured quantities of U, Th and Ra. Inter-session standard and  
148 duplicate measurements enabled estimation of the analytical uncertainty in the calculated U-  
149 Th-Ra radioactive activity ratios. The analytical uncertainty is 0.2 % for ( $^{234}\text{U}/^{238}\text{U}$ ), 1.5 % for  
150 ( $^{230}\text{Th}/^{234}\text{U}$ ) and 1.7 % for ( $^{226}\text{Ra}/^{230}\text{Th}$ ) activity ratios.

### 151 **3.3 In situ $^{10}\text{Be}$ isotope**

152 For the in situ  $^{10}\text{Be}$  analysis, each subsample consists of 150 g of 250-500  $\mu\text{m}$  sieved powder  
153 obtained from quartering the 2 mm crushed bulk samples. Oxides and organic matter were  
154 removed by HCl leaching, and a sequential HF dissolution was performed to remove the  
155 meteoric  $^{10}\text{Be}$  and obtain purified quartz (e.g., Nishiizumi et al., 1989). After addition of a  $^9\text{Be}$   
156 carrier, the purified quartz was dissolved, and beryllium separation and purification were  
157 performed by ion exchange chromatography. After hydroxylation and targeting, the  $^{10}\text{Be}/^9\text{Be}$   
158 ratios were measured by accelerator mass spectrometry at ASTER (CEREGE, University of Aix-  
159 Marseille, France). The  $^{10}\text{Be}/^9\text{Be}$  ratios of the blanks and samples are approximately  $1 \times 10^{-15}$   
160 and  $1 \times 10^{-13}$ , respectively. The overall analytical uncertainty is <5 % for the calculated  $^{10}\text{Be}$   
161 concentrations (Table 3).

## 162 **4- Determination of the regolith production rate, denudation rate and chemical mobility**

### 163 **4.1 Regolith production rate from U-Th-Ra disequilibria**

164 The principle of determination of the regolith production rate from the analysis of U-Th-Ra  
165 disequilibria has been detailed in previous studies (e.g., Dosseto et al., 2008, 2012; Ma et al.,  
166 2010; Chabaux et al., 2011, 2013). The approach is based on the analysis of samples  
167 collected along the main weathering direction, in the present case the vertical direction. For

168 samples in the weathering zone, as the time variation of nuclide mobility during water rock  
 169 interactions is not known *a priori*, the time evolution of radioactive nuclides, is classically  
 170 described in terms of continuous processes involving, in addition to the radioactive decay  
 171 laws, loss and gain processes represented by first-order kinetic rate laws for the loss  
 172 processes and zero-order kinetic rate laws for the gain processes. For the  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$ -  
 173  $^{226}\text{Ra}$  system, this leads to the following equations:

$$\frac{d \text{}^{238}\text{U}}{dt} = f_{238} \text{}^{238}\text{U}_0 - k_{238} \text{}^{238}\text{U} - \lambda_{238} \text{}^{238}\text{U} \quad (1)$$

$$\frac{d \text{}^{234}\text{U}}{dt} = f_{234} \text{}^{234}\text{U}_0 + \lambda_{238} \text{}^{238}\text{U} - k_{234} \text{}^{234}\text{U} - \lambda_{234} \text{}^{234}\text{U} \quad (2)$$

$$\frac{d \text{}^{230}\text{Th}}{dt} = f_{230} \text{}^{230}\text{Th}_0 + \lambda_{234} \text{}^{234}\text{U} - k_{230} \text{}^{230}\text{Th} - \lambda_{230} \text{}^{230}\text{Th} \quad (3)$$

$$\frac{d \text{}^{226}\text{Ra}}{dt} = f_{226} \text{}^{226}\text{Ra}_0 + \lambda_{230} \text{}^{230}\text{Th} - k_{226} \text{}^{226}\text{Ra} - \lambda_{226} \text{}^{226}\text{Ra} \quad (4)$$

174 where  $\lambda_i$ ,  $k_i$  and  $f_i$  are the radioactive decay, loss and gain constants (in  $\text{yr}^{-1}$ ), respectively, for  
 175 the radionuclides  $i$  (i.e.,  $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{230}\text{Th}$  and  $^{226}\text{Ra}$ ). In the equations,  $t$  is the time elapsed  
 176 between the reference weathering state and its current state. For simplification during the  
 177 equation-solving procedure, the input fluxes are expressed as a proportion of the number of  
 178 atoms of nuclides added per year to the initial sample. The loss ( $k$ ) and gain ( $f$ ) terms used in  
 179 the equations are usually assumed to be constant with time for the purpose of simplicity and  
 180 tractability (e.g., Dequincey et al., 2002; Dosseto et al., 2008; Ma et al., 2010; Chabaux et al.,  
 181 2013). The mean production rate of regolith,  $P$  (in  $\text{mm/kyr}$ ), can be estimated through the  
 182 resolution of the above equation system and the determination of the age  $\Delta t$  (in kyr) of a  
 183 sample relative to a reference sample from deeper in the profile at a distance  $\Delta h$  (in mm)

184 along the weathering direction. For isovolumetric weathering, P is given by the following  
185 equation:

$$P = \frac{\Delta h}{\Delta t} \quad (5)$$

186

187 As the gain and loss coefficients in equations 1-4 are not known and the  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$ - $^{226}\text{Ra}$   
188 analyses yield only three independent data per sample, namely the ( $^{234}\text{U}/^{238}\text{U}$ ), ( $^{230}\text{Th}/^{234}\text{U}$ )  
189 and ( $^{226}\text{Ra}/^{230}\text{Th}$ ) ratios, the analysis of only one sample (in addition to a reference sample) is  
190 not sufficient to determine the weathering production rate P. The mathematical formalism  
191 developed to retrieve such time information assumes that the gain and loss coefficients ( $k_i$   
192 and  $f_i$ ) of the model can be considered constants, at least for a part of the profile from which  
193 several different samples can be collected and analyzed. The measured ( $^{234}\text{U}/^{238}\text{U}$ ),  
194 ( $^{230}\text{Th}/^{234}\text{U}$ ) and ( $^{226}\text{Ra}/^{230}\text{Th}$ ) ratios of each sample are used to determine the mobility  
195 parameters of the model and the age of different samples relative to the reference sample.  
196 The numerical solutions are obtained using a stochastic quantum particle swarm  
197 optimization scheme (more details in Chabaux et al., 2013), which minimizes the sum of the  
198 squared differences between the measured and estimated activity ratios. The parameter  
199 solutions that allow the model to fit the observed activity ratios within 1 % error are  
200 retained and averaged to obtain a set of optimized output parameter values. The  
201 uncertainties in the parameter estimates are calculated as two-sigma standard deviations on  
202 the mean parameter estimate. It is important to stress here that such modeling approaches  
203 only give mean long-term values, i.e., over several kyr or 10 kyr, for regolith production rates  
204 and nuclide gain and loss parameters.

#### 205 **4.2 Regolith denudation rate from in situ $^{10}\text{Be}$ depth profile**

206 As with U-series disequilibrium methodology for regolith production rate, the depth  
 207 variations of in situ <sup>10</sup>Be concentrations within a weathering profile are recognized as a  
 208 powerful approach to constrain geomorphologic parameters, including the mean  
 209 denudation rate of a regolith (e.g., Siame et al., 2004; Dosseto and Schaller, 2016). In situ  
 210 <sup>10</sup>Be is produced at the surface of the Earth by the interaction of cosmic rays and rock atomic  
 211 nuclei (Brown et al., 1995). By taking into account (1) the different attenuation lengths of  
 212 secondary neutrons and muons, which produce <sup>10</sup>Be mainly in surface horizons for neutrons  
 213 and at greater depths for muons (Kim and Englert, 2004), and (2) their relative contribution  
 214 to the total produced <sup>10</sup>Be, the depth variations of in situ <sup>10</sup>Be concentrations within a  
 215 weathering profile is given by (e.g., Siame et al., 2004; Braucher et al., 2009):

$$\begin{aligned}
 C(t, d, z) = & C_0 \exp(-\lambda t) + \frac{P_0 * P_n}{\frac{\rho d}{K_n} + \lambda} \exp\left(\frac{-\rho z}{K_n}\right) \left(1 - \exp\left(-t\left(\frac{\rho d}{K_n} + \lambda\right)\right)\right) \\
 & + \frac{P_0 * P_{nm}}{\frac{\rho d}{K_{nm}} + \lambda} \exp\left(\frac{-\rho z}{K_{nm}}\right) \left(1 - \exp\left(-t\left(\frac{\rho d}{K_{nm}} + \lambda\right)\right)\right) \\
 & + \frac{P_0 * P_{fm}}{\frac{\rho d}{K_{fm}} + \lambda} \exp\left(\frac{-\rho z}{K_{fm}}\right) \left(1 - \exp\left(-t\left(\frac{\rho d}{K_{fm}} + \lambda\right)\right)\right) \quad (6)
 \end{aligned}$$

216 where  $t$  is the exposure age of the profile (in yr),  $d$  is the mean denudation rate (in cm/yr),  $z$   
 217 is the depth of each sample (in cm),  $C_0$  is the initial concentration from previous exposure (in  
 218 atoms/g quartz),  $\lambda$  is the radioactive decay of the <sup>10</sup>Be (in yr<sup>-1</sup>) and  $\rho$  is the integrated density  
 219 of each sample (in g/cm<sup>3</sup>).  $P_0$  is the total production rate of <sup>10</sup>Be at the surface of the soil  
 220 (12.17 at/g/yr and 0.266 at/g/yr for spallation and muons, respectively, determined with the  
 221 CRONUS online calculator; Balco et al., 2008).  $P_n$ ,  $P_{nm}$  and  $P_{fm}$  are the relative contributions  
 222 of 97.85, 1.5 and 0.65 % to the total production and  $K_n$ ,  $K_{nm}$  and  $K_{fm}$  are the effective

223 attenuation lengths of 150, 1500 and 5300 g/cm<sup>2</sup>, for secondary neutrons, negative muons  
224 and fast muons, respectively (Braucher et al., 2009). The depth variations of in situ <sup>10</sup>Be  
225 concentrations in a weathering profile can thus be used to estimate the exposure age and  
226 the mean denudation rate of a regolith, in addition to the inherited <sup>10</sup>Be concentration,  
227 without any steady state assumption.

### 228 **4.3 Chemical mobility and volumetric variation**

229 In addition to the determination of regolith production and denudation rates from the above  
230 approaches, the chemical mobility and volumetric variation associated with the weathering  
231 processes can be estimated through the analysis of major and trace element concentrations.  
232 Based on the presence of an immobile element *j*, the chemical mobility of an element *i*  
233 during weathering can be quantified by mass transfer coefficients (Brimhall et al., 1991), as  
234 defined by the following:

$$\tau_i = \left( \frac{C(i)_{sample} * C(j)_{bedrock}}{C(i)_{bedrock} * C(j)_{sample}} - 1 \right) \quad (7)$$

235  
236 where  $C(i)_{sample}$  and  $C(i)_{bedrock}$  represent the concentrations of the element *i* for a given  
237 sample and for the deepest bedrock sample and  $C(j)_{sample}$  and  $C(j)_{bedrock}$  represent the  
238 concentrations of the immobile element *j* for a given sample and for the deepest bedrock  
239 sample, respectively. Positive or negative  $\tau_i$  values imply the gain or loss, respectively, of the  
240 element *i* relative to the deepest bedrock sample, taken here as the reference rock. In  
241 addition, the chemical depletion fraction (Riebe et al., 2003), allows an estimation of the  
242 denudation fraction due to chemical weathering between the reference bedrock sample and  
243 a given sample and is defined by the following:

$$CDF = \left( 1 - \frac{C(j)_{bedrock}}{C(j)_{sample}} \right) \quad (8)$$

244 It is also possible to estimate the relative volume variation associated with rock weathering  
 245 between two considered levels by the volumetric strain index calculation (Brimhall et al.,  
 246 1991; Riebe et al., 2003):

247

$$\varepsilon = \left( \frac{C(j)_{bedrock} * d_{bedrock}}{C(j)_{sample} * d_{sample}} \right) - 1 \quad (9)$$

248

249 where  $d_{bedrock}$  and  $d_{sample}$  are the bulk density values of the bedrock and the considered  
 250 sample, respectively. Values of  $\varepsilon$  close to zero imply isovolumetric weathering, while positive  
 251 or negative values indicate expansion or compaction, respectively.

## 252 **5- Results**

### 253 **5.1 Mineralogical and geochemical variations within the weathering profile**

254 The whole rock XRD analyses are presented in Appendix A and in Table EA1. These data  
 255 along with the thin section observations are consistent with previously published data for  
 256 the Strengbach site (e.g., Fichter et al., 1998) and indicate that quartz and muscovite do not  
 257 present any significant evidence of weathering in the granitic bedrock. Biotite is weathered  
 258 at all depths and replaced by clays, sericite and hematite. The K-feldspar and plagioclase  
 259 crystals show structural and mineralogical evolution from the base to the top of the granite,  
 260 with a higher density of clay filled cracks in samples from the granite top. Carlsbad and  
 261 polysynthetic twins are identifiable at the granite base, while it is more difficult to recognize  
 262 them at the granite top. The analysis of the <2  $\mu\text{m}$  clay fraction (Table EA1) indicates that the

263 clay composition is relatively homogeneous within the granitic bedrock and is dominated by  
264 illite (40 to 60 % illite, 20 to 30 % inter-stratified illite/smectite, 5 to 25 % of smectite and 5  
265 to 10 % of kaolinite). The clay composition is different and more variable in the soil, where  
266 the smectite and inter-stratified clays become dominant (60 to 70 %) followed by the illite  
267 (20 to 30 %) and kaolinite (5 to 10 %). These data show a significant increase in supergene  
268 alteration from the base to the top of the profile, in accordance with the increase in the loss  
269 on ignition (LOI, Table 1).

270 The variations in the major and trace element concentrations within the weathering profile  
271 (Table 1) have been used to estimate the chemical mobility associated with weathering by  
272 the calculation of the mass transfer coefficients. For this calculation, Ti is often considered  
273 one of the best-suited immobile elements (e.g., Egli et al., 2006). For the tau calculation, the  
274 normalization to Ti rather than to Zr concentration has been preferred, even if Zr can be  
275 considered immobile in such environment (Rihs et al., 2011). The reason is the better  
276 analytical precision obtained for the Ti concentrations (3%) than for Zr ( $\approx 10\%$ ), which leads  
277 to a better precision of the tau value determined with Ti ( $\approx 10\%$ ) than with Zr ( $\approx 20\%$ ). The  
278 calculation of relatively constant ratios within the profile between Ti and other elements  
279 usually considered immobile, such as Ta (not shown) and Th (Figure 2c), especially in the  
280 deeper part of the profile, reinforces the assumption that Ti is mainly immobile during  
281 weathering. Tau values indicate no significant mobility for the main major elements ( $\text{Na}_2\text{O}$ ,  
282  $\text{K}_2\text{O}$ ,  $\text{CaO}$ ,  $\text{Al}_2\text{O}_3$ , and  $\text{SiO}_2$ ) in the deeper part of the granitic profile below 100 cm depth  
283 (Figure 2a). The depletion becomes significant in the regolith portion above 100 cm depth,  
284 with chemical mobility trends that differ from one element to another, especially in the  
285 uppermost part of the soil known to be significantly affected by biological processes,  
286 vegetation cycling and atmospheric deposition (e.g., Stille et al., 2011; Gangloff et al., 2014).

287 In this upper part of the profile, the volumetric strain index indicates a significant expansion  
288 of the soil horizons, with  $\varepsilon$  values from 50 to 150 %, while deeper in the profile, in the  
289 granite and saprolite horizons, the weathering is nearly isovolumetric (Figure 3b).

290 As for tau values, the depth variations in the U/Th concentration ratios measured by isotopic  
291 dilution, which are more precisely estimated than the tau values, allow for subdividing the  
292 profile into two main zones, with U loss in the lower part of the profile, which gradually  
293 increases upwards up to approximately a depth of 100 cm, and a U/Th ratio increase toward  
294 the surface above 100 cm (Figure 2b). This increase might indicate Th migration due to  
295 complexation with organic colloids, as observed at other sites (Rihs et al., 2011), or U inputs  
296 from external atmospheric deposition, as suggested in other contexts (Pelt et al., 2013;  
297 Chabaux et al., 2013). Regardless of the precise origin of this increase, the U/Th data confirm  
298 that the transition between granitic bedrock and regolith at a depth of approximately 100  
299 cm vertically divides the weathering profile into two distinct zones that are marked by  
300 different chemical mobility values and likely exhibit specific weathering processes.

## 301 **5.2 U-Th-Ra and in situ <sup>10</sup>Be systematics**

302 The depth subdivision of the profile inferred from major and trace element mobility is also  
303 observed in the U-Th-Ra disequilibrium data (Table 2, Figure 4). The lower part of the profile  
304 (below 100 cm depth) is characterized by rather simple and continuous variation trends in  
305 the (<sup>234</sup>U/<sup>238</sup>U), (<sup>230</sup>Th/<sup>234</sup>U) and (<sup>226</sup>Ra/<sup>230</sup>Th) activity ratios with depth, with an increase in  
306 the (<sup>234</sup>U/<sup>238</sup>U) and (<sup>230</sup>Th/<sup>234</sup>U) ratios with decreasing depth and a decrease in the  
307 (<sup>226</sup>Ra/<sup>230</sup>Th) ratios with decreasing depth (Figure 4). In the upper regolith, the (<sup>234</sup>U/<sup>238</sup>U)  
308 and (<sup>230</sup>Th/<sup>234</sup>U) activity ratios define clearly opposite trends with depth, which support the  
309 suggestion that the two parts of the profile are marked by different weathering processes.

310 The upper part of the profile is also characterized by much more scattered ( $^{226}\text{Ra}/^{230}\text{Th}$ )  
311 ratios, without a simple correlation with depth.

312 For in situ  $^{10}\text{Be}$ , the concentrations point to a progressive decrease with depth but with two  
313 different patterns of variation in the profile, one in the deeper horizons, where the  
314 weathering is isovolumetric, and the second one in the uppermost regolith horizons (<50  
315 cm), which are affected by volumetric expansion (Table 3, Figure 3). The deeper part is  
316 characterized by a common exponential decrease in  $^{10}\text{Be}$ , while the soil to upper regolith  
317 part (0-50 cm) is characterized by a linear decrease in the in situ  $^{10}\text{Be}$  concentration with  
318 depth (Figure 3).

## 319 **6. Discussion**

### 320 **6.1 Determination of regolith production and denudation rates**

321 The mineralogical and geochemical data lead to a depth-dependent division of the profile  
322 into two main parts, with progressive variation trends of the U-series systematic in the  
323 bedrock and more scattered variations in the upper horizons (Fig. 4). Similar to the results of  
324 the analyses of the short half-life  $^{228}\text{Ra}$  and  $^{228}\text{Th}$  nuclides in soil profiles from different forest  
325 catchments, including the Strengbach (Rihs et al., 2011; Gontier et al., 2015), the short-  
326 distance variations in the upper horizons most likely result from preferential Th mobility  
327 related to the differential mobilization of radionuclides in the presence of organic matter. As  
328 also emphasized in Gontier et al., (2015), such short-distance redistributions in the soil  
329 horizons may imply significant variability of the mobility coefficients in this part of the  
330 profile, at least for Th isotopes. Thus, this effect prevents the direct application of the  
331 radioactive disequilibrium methodology described in section 4-1. In the Strengbach case, and  
332 likely in many other forest contexts, the bedrock is thus the most suitable place to quantify

333 the mean long-term regolith production rate via the conventional U-Th-Ra methodology. In  
334 the present study, the modeling approach has been therefore applied only to the lower part  
335 of the profile, in which Th can be assumed to be immobile (Figure 2c), consequently reducing  
336 the number of unknowns in the modeling ( $k_{230}=0$  and  $f_{230}=0$ ). The modeling results (Figure 4)  
337 show that the mean U mobility parameters fall within the range of existing data for shale  
338 and granodiorite lithologies (Ma et al., 2010; Dequincey et al., 2002; Dosseto et al, 2008;  
339 Chabaux et al, 2013). As evidenced by the obtained coefficients, the particular positive  
340 correlation between  $(^{234}\text{U}/^{238}\text{U})$  and  $(^{230}\text{Th}/^{234}\text{U})$  ratios in the deeper part of the profile do  
341 not correspond to a simple leaching of U. To explain the ratios of  $(^{234}\text{U}/^{238}\text{U})>1$  and  
342  $(^{230}\text{Th}/^{234}\text{U})>1$ , U removal must have occurred at a higher rate than U input, and incoming U,  
343 which may originate from U mobilization in the upper horizons, must have been  
344 characterized by a higher  $^{234}\text{U}$ - $^{238}\text{U}$  fractionation than the removed U (Figure 4, details also in  
345 Dequincey et al., 2002). Such mobility implies precipitation of U-rich secondary minerals,  
346 such as Fe-oxyhydroxides, from fluids with ratios of  $(^{234}\text{U}/^{238}\text{U})>1$ . This hypothesis is  
347 supported by the ratios of  $(^{234}\text{U}/^{238}\text{U})>1$  in soil solutions from a neighboring profile in the  
348 same part of the watershed (Prunier et al., 2015). The U-Th-Ra modeling also gives the  
349 elapsed time between the granite base and the lower regolith, which yields a mean regolith  
350 production rate of  $12\pm 4$  mm/kyr. Considering the density of 2.5 for the lower regolith, the  
351 regolith production rate in terms of specific flux is  $30\pm 10$  T/km<sup>2</sup>/yr.

352 For application of the in situ <sup>10</sup>Be approach (section 4-2), a numerical inverse procedure has  
353 been used to determine the exposure age, the mean denudation rate and the inherited  
354 concentration that best explain the data from equation 6. The inverse procedure consists of  
355 minimizing the chi-square value of the system by the algorithm of Levenberg-Marquardt

356 (Marquardt, 1963) and has been adapted to take into account the volumetric expansion of  
357 the upper regolith. The chi-square value is defined as follows:

$$X = \sum \left( \frac{C_{data}(z) - (C_{model}(t, d, C_0, z) + \Delta C)}{\sigma(z)} \right)^2 \quad (10)$$

358

359 where  $C_{data}(z)$  and  $C_{model}(t, d, C_0, z)$  are the measured and modeled  $^{10}\text{Be}$  concentrations,  
360 respectively, and  $\sigma(z)$  is the uncertainty in the measured  $^{10}\text{Be}$  concentration for a sample at  
361 depth  $z$ . A correction factor  $\Delta C$  has been introduced to take into account the volumetric  
362 expansion of the upper 50 cm because the density variation only is insufficient to explain the  
363 concentration trend of this part of the profile. This implies that expansion must have  
364 occurred relatively late (<5–10 kyr) in the history of the profile, as the measured  
365 concentrations do not reflect the corresponding production rate changes. Uncertainty in the  
366 parameters is estimated from the covariance matrix of the problem. It is important to stress  
367 that, considering the mean denudation and the surface  $^{10}\text{Be}$  production rates at the summit  
368 of the Strengbach watershed, the current accumulation of  $^{10}\text{Be}$  within the profile is not  
369 balanced by erosion and radioactive decay. Because a  $^{10}\text{Be}$  concentration steady state is not  
370 present, the geochronometric application of the modeling is possible. The best fit gives an  
371 exposure age of the profile of  $19,734 \pm 2,168$  yr, a mean denudation rate of  $22 \pm 10$  mm/kyr (in  
372 terms of specific flux,  $37 \pm 15$  T/km<sup>2</sup>/yr, based on the mean density of the regolith of 1.7) and  
373 an inherited  $^{10}\text{Be}$  concentration of  $15,000 \pm 1,000$  at/g quartz (Figure 3).

## 374 **6.2 Implications for regolith evolution on the millennial timescale**

375 To the best of our knowledge, this study is likely the first one to combine U-series and in situ  
376  $^{10}\text{Be}$  analyses on the same samples in a single weathering profile with such a depth extent

377 and spatial resolution. This coupled approach allows for determining, in addition to the  
378 regolith denudation and production rates, the exposure age of the profile and the potential  
379  $^{10}\text{Be}$  inherited concentration. As illustrated in the following, these estimates are relevant for  
380 recovering the Quaternary dynamics of the weathering profile.

381 The determined exposure age inferred from the  $^{10}\text{Be}$  data indicates the occurrence of an  
382 erosive event at the summit of the Strengbach watershed approximately 20,000 yr ago, likely  
383 resulting from glacial or peri-glacial processes in relation with the last Würm glaciation,  
384 which is in accordance with regional investigations (Seret et al., 1990; Mercier et al., 1999).

385 In addition, the occurrence of an inherited  $^{10}\text{Be}$  concentration within the bedrock indicates a  
386 complex erosive history at the summit of the Strengbach catchment. By extrapolating the  
387 long-term regolith propagation rate determined from U-Th-Ra variations (i.e.,  $30\pm 10$   
388  $\text{T}/\text{km}^2/\text{yr}$  or  $12\pm 4$   $\text{mm}/\text{kyr}$ ), an age of approximately 150 kyr is obtained for the entire 2 m-  
389 thick profile analyzed in this study. This age would lead to a  $^{10}\text{Be}$  concentration at 2 m depth  
390 much higher than the inherited  $^{10}\text{Be}$  concentration of 15,000 at/g quartz obtained from our  
391 data. In this bedrock, the measured inherited  $^{10}\text{Be}$  concentration corresponds to the  
392 concentration obtained from a muonic exposure at approximately 4 m depth over  $\approx 150$  kyr.

393 A way to reconcile all the data, assuming the above determined average denudation rate of  
394 22  $\text{mm}/\text{kyr}$ , is that a thickness of 2 m was eroded 20 kyr ago and that a total resetting of the  
395 bedrock  $^{10}\text{Be}$  inventory must have occurred 150 kyr ago, which is consistent with the fact  
396 that the penultimate Riss glaciation was probably more intense than the last Würm  
397 glaciation in the mountains of central Europe (Dehnert et al., 2010).

398 The independent determination of regolith production and denudation rates from U-Th-Ra  
399 isotopes and in situ  $^{10}\text{Be}$  constitutes an interesting method for discussing the stability state

400 of a weathering profile on a millennial timescale. For the Strengbach case, the proposed  
401 approach gives similar values for denudation rate ( $37\pm 15$  T/km<sup>2</sup>/yr) and production rate  
402 ( $30\pm 10$  T/km<sup>2</sup>/yr), suggesting that at a millennial time scale, the regolith mass balance at the  
403 summit of the catchment is close to steady state, even if it was perturbed 20 kyr ago. Such a  
404 conclusion may indicate that the return to mass balance steady state is relatively fast in this  
405 system, i.e., obtained in less than 20 kyr and/or that the erosive event that occurred 20 kyr  
406 ago has only very transiently disturbed the processes controlling regolith formation at the  
407 summit of the Strengbach watershed. The results may also suggest that the recent  
408 anthropogenic perturbations linked to the 20<sup>th</sup> century, e.g., acid rain or the recent  
409 afforestation efforts, which may have recently modified the cationic concentrations and  
410 acidity of soil and stream waters in the watershed (Viville et al., 2012; Prunier et al., 2015),  
411 have not perturbed the <sup>10</sup>Be and U-Th-Ra long-term records.

412 Very few other studies combining U-series disequilibrium and cosmonuclides methodology  
413 exist for other sites, even when including those involving the meteoritic <sup>10</sup>Be approach  
414 (Table 4). In addition, these other studies usually dealt with more superficial regolith and  
415 different sample sets for isotopic analyses. However, despite these limitations, the data  
416 indicate that in all the investigated mid-mountain catchments, which are characterized by  
417 different climatic and tectonic settings, there is a fairly good consistency between the long-  
418 term regolith production and denudation rates (Table 4). This seems to be especially true at  
419 the ridge-tops and/or hill-crests of watersheds, where geochronological interpretations are  
420 likely less affected by colluvial transport and deposition. Altogether, even if such a  
421 conclusion remains preliminary, the current data suggest that the long-term regolith  
422 evolution could be commonly at steady state.

### 423 **6.3 Comparison of millennial and present-day weathering and erosion rates**

424 Because U-series and  $^{10}\text{Be}$  approaches integrate long-term processes, a substantial step to  
425 understand the recent regolith evolution could be provided by the comparison of long-term  
426 weathering and erosion rates with present-day estimates. As established by Riebe et al.  
427 (2003), the knowledge of denudation rates determined by cosmogenic nuclide, combined  
428 with the geochemical mass balance approaches and the definition of the chemical depletion  
429 fraction (CDF), allow to distinguish the chemical and physical components of the total  
430 denudation. This approach is predicated on an assumption of steady state, which is  
431 supported by the consistency between U-series and  $^{10}\text{Be}$  results. The total weathering rate in  
432  $\text{T}/\text{km}^2/\text{yr}$  is as follows:

$$W = D \left( 1 - \frac{C(\text{Ti})_{\text{rock}}}{C(\text{Ti})_{\text{soil}}} \right) = D \times CDF \quad (11)$$

433  
434 where  $D$  is the mean denudation rate in  $\text{T}/\text{km}^2/\text{yr}$  inferred from the cosmogenic  $^{10}\text{Be}$  and  
435  $C(\text{Ti})_{\text{rock}}$  and  $C(\text{Ti})_{\text{soil}}$  are the concentrations of titanium (considered an immobile  
436 element) in the bedrock and in the soil, respectively. The application of this approach to the  
437 bedrock and the soil levels of the studied weathering profile leads to a CDF value of 0.22  
438 when using for the average Ti concentrations of the bedrock and soil levels. A maximum CDF  
439 value of 0.30 is obtained by using the lowest Ti concentrations measured in the bedrock and  
440 the highest concentrations in the soil. Similar calculations can be made with Zr, which can  
441 also be considered an immobile element in the Strengbach catchment (Rihs et al., 2011):  
442 CDF values between 0.13 and 0.25 are obtained according to whether the average  
443 concentration values or the extreme values are used for each level. These different

444 calculations indicate that at the profile scale, long-term chemical weathering accounts for 13  
445 to 30 % of the total denudation, which yields a long-term chemical weathering flux of  
446 between 5 and 11 T/km<sup>2</sup>/yr and consequently a long-term erosion flux of between 26 and 32  
447 T/km<sup>2</sup>/yr. Therefore, physical erosion is the main process controlling the loss of regolith.

448 The above weathering flux calculations can be easily adapted to calculate the long-term  
449 weathering flux of a specific chemical element by using the mean denudation rate ( $D$ ), the  
450 elemental concentrations within the bedrock ( $C(x)_{bedrock}$ ) and the tau values ( $\tau_x$ ) in the  
451 upper soil (Riebe et al., 2003):

$$W_x = D * C(x)_{bedrock} * (-\tau_x) \quad (12)$$

452

453 Based on this approach, long-term weathering fluxes of approximately  $3 \pm 2$  T/km<sup>2</sup>/yr for Si  
454 and  $0.7 \pm 0.47$  T/km<sup>2</sup>/yr for cations can be estimated from the geochemical variations along  
455 the regolith profile at the watershed summit. Similarly, the regular analyses of the dissolved  
456 fluxes and the sediment loads carried by the Strengbach stream allow for estimation of the  
457 current erosion and weathering rates at the watershed scale. The combination of the  
458 suspended and bed load monitoring yields a total erosion rate of  $5 \pm 1$  T/km<sup>2</sup>/yr at the  
459 watershed outlet (Viville et al., 2012). By correcting the net flux for basic cations from  
460 atmospheric deposition, a cationic weathering flux of  $1.98 \pm 0.2$  T/km<sup>2</sup>/yr and a Si  
461 weathering flux of  $2.89 \pm 0.3$  T/km<sup>2</sup>/yr are estimated (Viville et al., 2012). The current  
462 exported flux for Si determined at the watershed outlet is surprisingly very close to the long-  
463 term Si weathering flux associated with regolith formation inferred for the watershed  
464 summit. This consistency means that the first-order chemical weathering, which is controlled  
465 by the rates of primary mineral dissolution and secondary mineral precipitation, is rather

466 spatially uniform and stable in time. In contrast, the current cationic flux in the Strengbach  
467 stream seems to be higher than the long-term flux determined at the profile scale for basic  
468 cations. This difference may be due to a recent increase in basic cation leaching from  
469 Strengbach soils, possibly triggered by the acidic atmospheric inputs and/or the forest  
470 management evolution experienced by the Strengbach catchment in recent decades (Viville  
471 et al., 2012; Prunier et al., 2015). In contrast to the weathering rates, the long-term erosion  
472 rate determined from  $^{10}\text{Be}$  and CDF data (approximately 30 T/km<sup>2</sup>/yr) is clearly higher than  
473 the current erosion rate determined in the Strengbach stream (approximately 5 T/km<sup>2</sup>/yr;  
474 Viville et al., 2012). The latter value is certainly an underestimate, as the suspended load was  
475 sampled only once every one or two weeks, which probably does not allow for a correct  
476 estimate of the suspended load exported during flood events. A more recent study of the  
477 erosion fluxes exported from the Strengbach watershed at its outlet, including an accurate  
478 determination of the suspended load exported from the watershed during flood events,  
479 yielded an erosion rate no higher than 10 T/km<sup>2</sup>/yr (Cotel et al., 2016). Regarding the  
480 sampling frequency developed in the latter study, and even if the bed load is always difficult  
481 to estimate precisely, an additional bias for the current erosion rates by a factor of three  
482 seems improbable. Thus, unlike weathering fluxes, the long-term erosion rate determined at  
483 the watershed summit and the present-day rate at the watershed scale are most likely  
484 different. Such a discrepancy is certainly not surprising, and can result from spatial  
485 heterogeneity in the erosion rate at the watershed scale, with high erosion rates along the  
486 main crests possibly counterbalanced by sediment storage downhill. It could also result from  
487 temporal variability in the physical erosion rate, with, for instance, high erosion rates during  
488 cold conditions, probably persisting long after the LGM, followed by lower erosion rates due

489 to stabilization of the regolith by soil and vegetation cover development during the  
490 Holocene.

491 As illustrated in the Strengbach case, this combination of U-series, in situ  $^{10}\text{Be}$  and stream  
492 monitoring allows for discussing the weathering and erosion variability at different spatial  
493 and temporal scales. Another site where a similar comparison is possible is the Rio Icacos  
494 watershed (Table 5 in Chabaux et al., 2013). Similar to the Strengbach catchment, an analysis  
495 suggests relatively similar long-term and short-term weathering rates, whereas significant  
496 differences exist between long-term denudation rates inferred from  $^{10}\text{Be}$  data and current  
497 erosion rates determined from sediment monitoring, which is also similar to the Strengbach  
498 catchment (Table 4). However, in this case, unlike the Strengbach catchment, the current  
499 erosion rates are higher than the long-term ones. This large variability in the physical erosion  
500 fluxes through time is also suggested in a few other studies. In the Susquehanna Shale Hills  
501 Observatory, it has been proposed that current erosion rates may be lower than previous  
502 conditions because deep sediments in the valley floor appear to retain a record of enhanced  
503 colluvial transport in the past (West et al., 2013). In Australia, longer sediment residence  
504 times have been proposed for the Murrumbidgee River during warm periods, in association  
505 with the development of vegetation cover and a decrease in erosion (in Dosseto and  
506 Schaller, 2016). Even if, once again, there are very few sites where long-term trends are  
507 compared to short-term trends, it seems that mid-mountain watersheds that experienced  
508 glacial or periglacial conditions during the LGM (Strengbach, SSHO or Australian upper  
509 watersheds) recorded a decrease in erosion rates with the onset of Holocene. In contrast, in  
510 the Rio Icacos watershed, characterized by warmer conditions and hence unaffected by  
511 periglacial processes, the current erosion rates are faster (Table 4). Thus, the above data  
512 support the assumption deduced from the Strengbach case that the erosion rates have been

513 more variable than the chemical weathering rates during the Quaternary. The data might  
514 also suggest a possible role of periglacial processes in the erosional response to Quaternary  
515 climatic variations.

## 516 **7 Conclusions**

517 The study performed on a weathering profile sampled at the summit of the Strengbach  
518 catchment illustrates the interesting results of the combined analyses of in situ  $^{10}\text{Be}$   
519 concentrations,  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$ - $^{226}\text{Ra}$  disequilibria and major and trace element  
520 concentrations. As summarized in Figure 5, this combination is a powerful methodology for  
521 recovering key parameters of the long-term regolith evolution. This approach suggests that  
522 the long-term regolith evolution in the Strengbach watershed is relatively close to steady  
523 state, despite recent anthropogenic perturbations. In addition, in regard to the mineralogical  
524 properties of the profile and implicit assumptions of the U-Th-Ra modeling, it appears that in  
525 this site (as well as in many other forest contexts), the bedrock is the most suitable place to  
526 quantify the regolith production rate using the U-Th-Ra methodology. The results also  
527 highlight the importance of having a sufficiently dense sampling strategy along the regolith  
528 depth profile to recover the Quaternary polyphase history of the regolith from the analysis  
529 and modeling of in situ  $^{10}\text{Be}$ . In combination with the above data, geochemical and mass  
530 balance calculations performed in the Strengbach stream and along the weathering profile  
531 allow for investigating the regolith evolution at different spatial and temporal scales. The  
532 results indicate that physical erosion is the dominant process of regolith loss in the  
533 Strengbach catchment and has probably varied much more than chemical weathering  
534 through time. Although very few sites with U-series, in situ  $^{10}\text{Be}$  and stream monitoring data  
535 are available for comparison, the current data suggest that (1) mass balance steady state of

536 regolith can be commonly achieved in soil mantled landscapes and (2) physical erosion has  
537 exhibited higher variability than chemical weathering over the last 10-150 kyr. The combined  
538 U-series and in situ  $^{10}\text{Be}$  analyses in high spatial resolution depth profiles could become a  
539 powerful approach in the future for providing new quantitative insights on weathering  
540 profile dynamics and regolith erosive history over Holocene and Late Pleistocene time  
541 periods. The association of these approaches with the monitoring of water chemistry and  
542 sediment fluxes at watershed outlets seems also important to correctly positioning the  
543 present-day erosion and weathering rates relative to the past rates recorded by the U-Th-Ra  
544 and in situ  $^{10}\text{Be}$  inventories.

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556 contribution.

#### 557 **Table and figure captions**

558 Table 1: Some chemical and physical characteristics of the studied weathering profile. Major and  
559 trace element concentrations were determined by conventional tetraborate alkaline fusion followed  
560 by mass spectrometry and atomic emission spectrometry measurements (ICP-MS and ICP-AES  
561 procedure) (uncertainty <5 % for major element and <10 % for trace element concentrations). STR13-  
562 53<sup>2</sup> is a dissolution powder duplicate. STR13-53<sup>3</sup> is a quartering duplicate that encompasses the  
563 whole crushing process.

564 Table 2: U-series data. U and Th concentrations are calculated from isotopic dilution. The  
565 uncertainties are at the  $2\sigma$  level: 0.2 % and 0.5 % for U and Th concentrations, 0.2 % for ( $^{234}\text{U}/^{238}\text{U}$ ), 1  
566 % for ( $^{230}\text{Th}/^{232}\text{Th}$ ), 1.5 % for ( $^{230}\text{Th}/^{234}\text{U}$ ) and 1.7 % for ( $^{226}\text{Ra}/^{230}\text{Th}$ ) activity ratios. STR13-53<sup>2</sup> is a  
567 dissolution powder duplicate. STR13-53<sup>3</sup> is a quartering duplicate that encompasses the whole  
568 crushing process. For the activity ratio calculation, the following radioactivity constants were used:  
569  $\lambda_{232}=4.948\times 10^{-11}\text{ yr}^{-1}$ ,  $\lambda_{230}=9.158\times 10^{-6}\text{ yr}^{-1}$ ,  $\lambda_{234}=2.826\times 10^{-6}\text{ yr}^{-1}$ ,  $\lambda_{238}=1.551\times 10^{-10}\text{ yr}^{-1}$ , and  $\lambda_{226}=4.335\times 10^{-4}\text{ yr}^{-1}$  (Akovali, 1996; Cheng et al., 2000).

571 Table 3: In situ  $^{10}\text{Be}$  data for the studied weathering profile. The elevation is 1147 m a.s.l. The latitude  
572 and longitude are  $48^{\circ}12'29.4''\text{N}$  and  $7^{\circ}11'30.3''\text{E}$ , respectively. There is no shielding correction due to  
573 the summit location of the profile. The surface production rates were determined with the CRONUS  
574 online calculator (Balco et al., 2008) and are 12.17 at/g/yr and 0.266 at/g/yr for spallation and  
575 muons, respectively. The total  $^9\text{Be}$  represents the carrier and natural  $^9\text{Be}$  of the samples (ICP-AES  
576 measurements). AMS analysis calibration was performed with a BeO NIST 4325 standard at ASTER.  
577  $^{10}\text{Be}/^9\text{Be}$  are blank corrected values. Uncertainties include AMS analysis, carrier mass and blank  
578 uncertainties.

579  
580 Table 4: Synthesis table of the studied sites that allow for U-series and  $^{10}\text{Be}$  results comparison.  
581 When rates are not provided in  $\text{T}/\text{km}^2/\text{yr}$ , conversions from  $\text{mm}/\text{kyr}$  to  $\text{T}/\text{km}^2/\text{yr}$  have been  
582 performed by assuming a regolith bulk mean density of  $1700\text{ kg}/\text{m}^3$ . (1) This study. (2) Cotel et al.,  
583 2016. (3) Ma et al., 2010. (4) West et al., 2013. (5) Jin et al., 2010. (6) Schoonejans et al., 2016. (7) Dosseto  
584 et al., 2008. (8) Heimsath et al., 2000. (9) Chabaux et al., 2013. (10) Brown et al., 1995. (11) McDowell and  
585 Asbury, 1994.  $^{10}\text{Be}$  denudation rates have been determined by in situ  $^{10}\text{Be}$  data, except for West et  
586 al., 2013, where meteoritic  $^{10}\text{Be}$  has been used.

587 Figure 1: a) Regional map of the Vosges massif and location of the Strengbach catchment. b) Contour  
588 of the Strengbach watershed. c) Topographic map of the Strengbach watershed and location of the

589 studied weathering profile (map from OHGE). d) Sampling of the weathering profile. The pedological  
590 observation led to division of the weathering profile into 3 zones: the soil from 0 to 50 cm, the  
591 saprolite from 50 to 100 cm and the granitic weathered bedrock from 100 to 200 cm. Each box in the  
592 column represents a collected sample. Red and blue dots indicate samples for which isotopic U-Th-Ra  
593 and in situ  $^{10}\text{Be}$  analyses have been performed, respectively. Green dots indicate samples for which  
594 thin sections have been performed.

595 Figure 2: a) Mass transfer coefficients calculated from equation 7. The uncertainty is calculated from  
596 elemental concentration uncertainty propagation and is <20 % (error bars show 20 % level). b) U/Th  
597 ratio. The U and Th concentrations are calculated from isotopic dilution. The uncertainty is 0.2 % for  
598 U, 0.5 % for Th and is within size point. c) Th/Ti ratio. The Ti and Th concentrations are calculated  
599 from alkaline fusion. The uncertainty is <15 % (error bars show 15 % level).

600 Figure 3: a) Bulk density calculated by combining the fine fraction density and the density of blocks  
601 with respect to the proportion of blocks in each horizon. For the bedrock and the blocks of granite, a  
602 constant density of  $2700 \text{ kg/m}^3$  is assumed. b) Volumetric strain index calculated from equation 9.  
603 Uncertainty is calculated from elemental concentration uncertainty propagation and is <10 % (error  
604 bars show 10 % level) c)  $^{10}\text{Be}$  depth profile. In blue: measured  $^{10}\text{Be}$  concentrations. Uncertainty is <5  
605 % (error bars show 5 % level). In red: best fit obtained from optimization by Levenberg-Marquardt  
606 algorithm. In grey: modeled  $^{10}\text{Be}$  concentrations without taking into account soil expansion.

607 Figure 4: ( $^{234}\text{U}/^{238}\text{U}$ ), ( $^{230}\text{Th}/^{234}\text{U}$ ) and ( $^{226}\text{Ra}/^{230}\text{Th}$ ) radioactive activity ratios along the weathering  
608 profile. In blue: experimental data. The uncertainties are at the  $2\sigma$  level: 0.2 % for ( $^{234}\text{U}/^{238}\text{U}$ ), 1.5 %  
609 for ( $^{230}\text{Th}/^{234}\text{U}$ ) and 1.7 % for ( $^{226}\text{Ra}/^{230}\text{Th}$ ) measured activity ratios. In red: best fit obtained from  
610 numerical optimization. Calculated coefficients:  $k_{238} = 4.81 * 10^{-6} \pm 3.68 * 10^{-7}$ ,  $k_{234} = 5.47 * 10^{-6} \pm 9.20 * 10^{-7}$ ,  
611  $k_{226} = 2.22 * 10^{-5} \pm 4.44 * 10^{-6}$ ,  $f_{238} = 1.63 * 10^{-6} \pm 4.00 * 10^{-7}$ ,  $f_{234} = 2.76 * 10^{-6} \pm 8.96 * 10^{-7}$   
612 and  $f_{226} = 6.61 * 10^{-6} \pm 2.20 * 10^{-6}$  ( $2\sigma$  level, expressed in  $\text{yr}^{-1}$ ).  
613 Calculated weathering age is approximately 69,000 yr for the modeled section.

614 Figure 5: Summary of the methodologies applied to the regolith profile at the Strengbach catchment  
615 summit. Combined approaches allow for constraining the long-term evolution of the profile with  
616 comparison of both regolith production and denudation rates.

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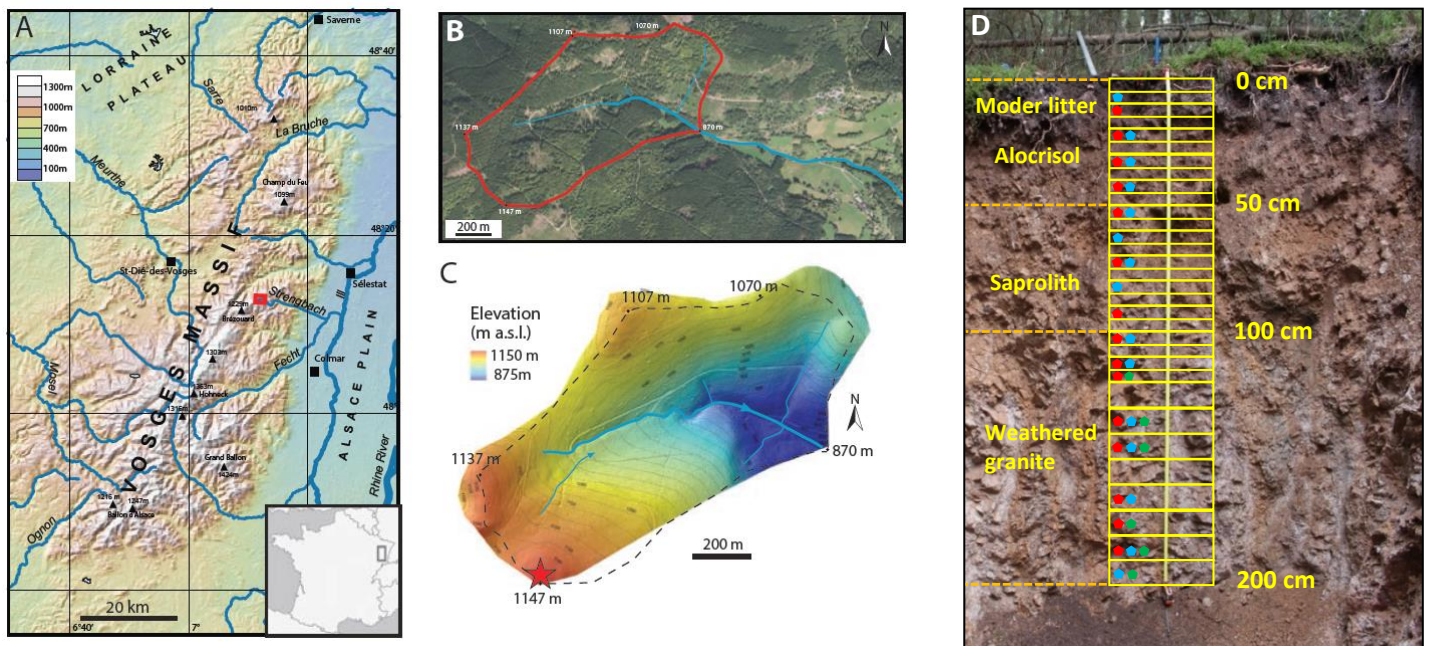


Figure 1

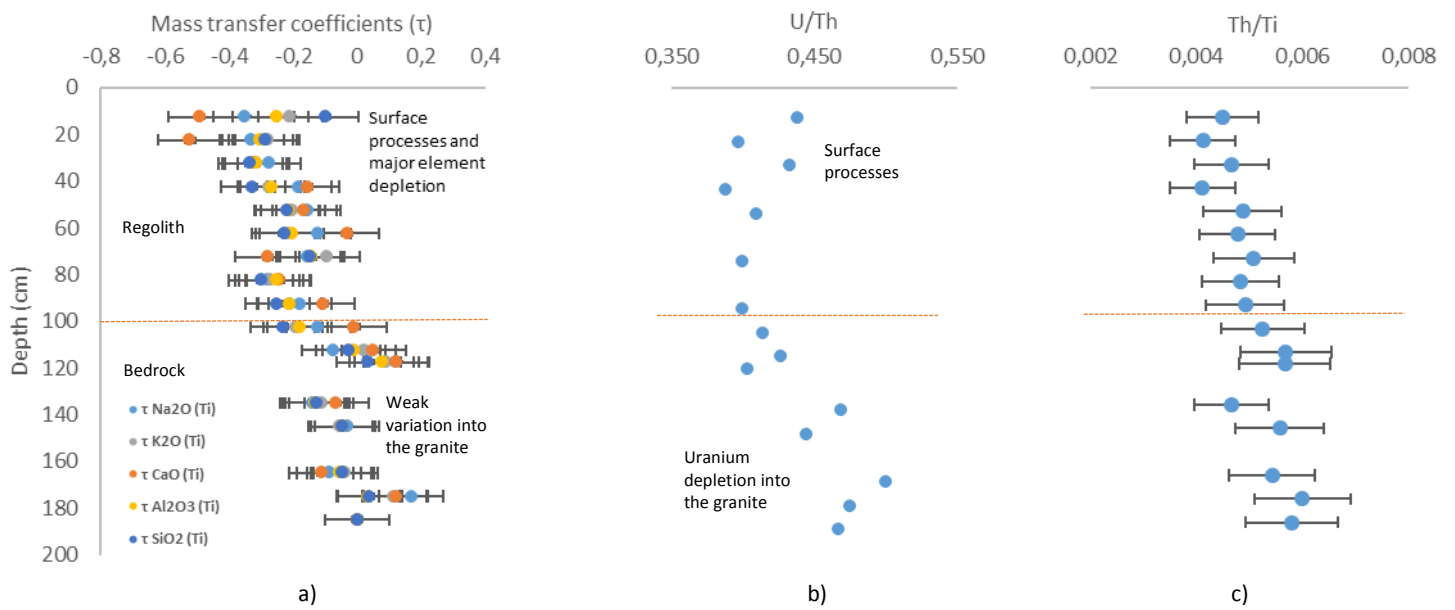


Figure 2

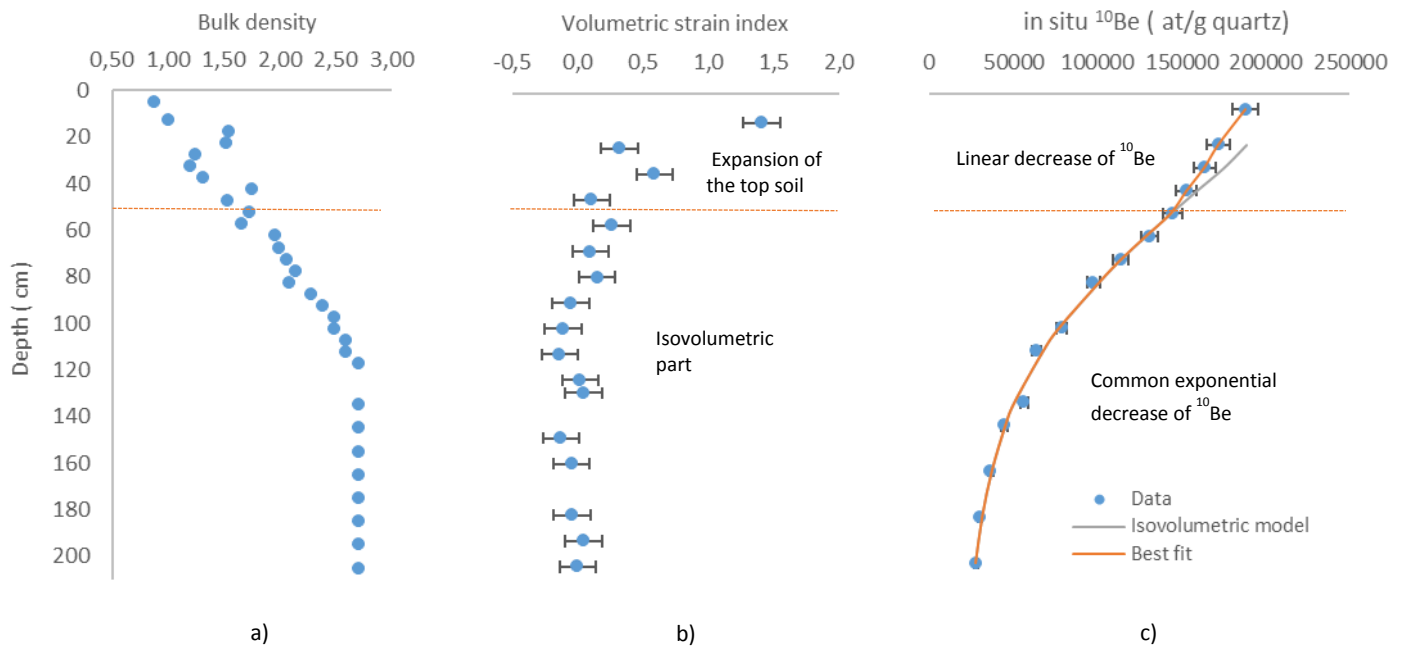


Figure 3

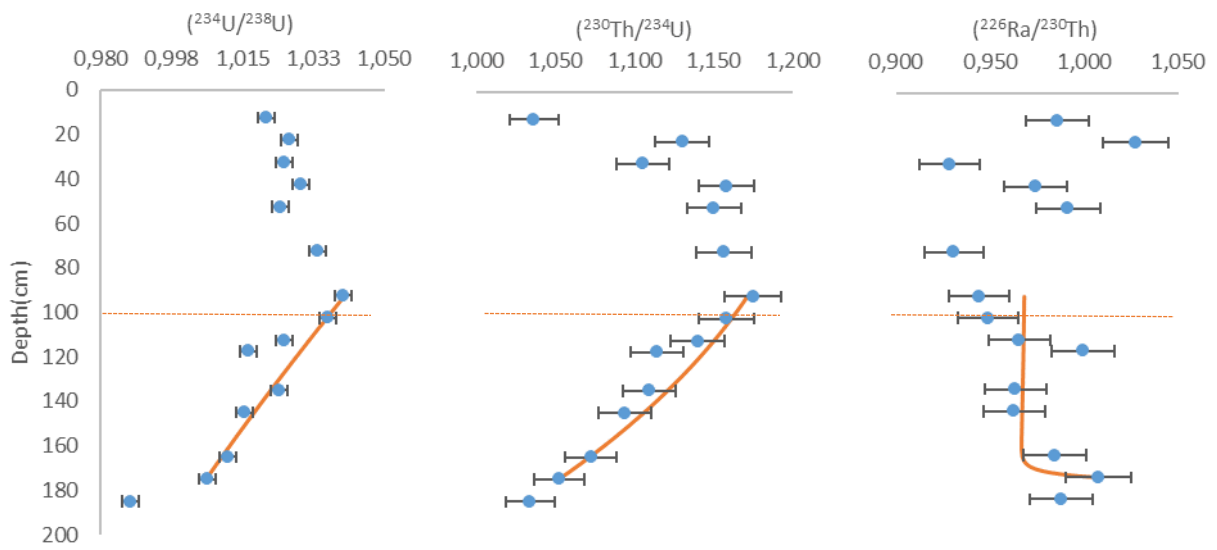


Figure 4

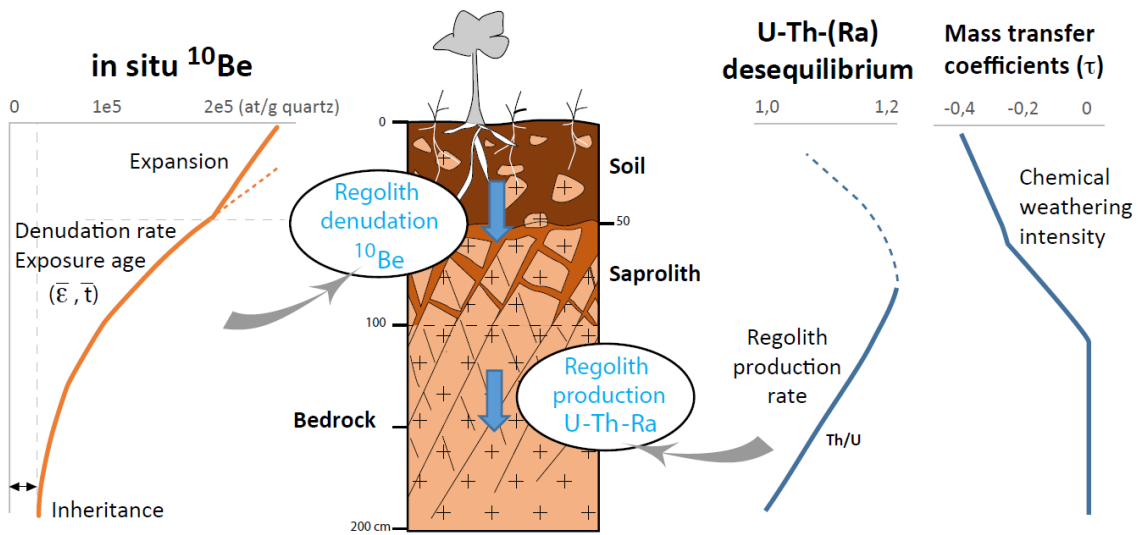


Figure 5

Sample	Depth (cm)	Quartz (%)	Plagioclase Feldspars (%)	K-feldspars (%)	Micas and illites (%)	Kaolinite (%)	Interstratified clays (%)	Hematite (%)
STR13-41	12.5	43	17	25	8	2	5	0
STR13-43	22.5	43	18	25	8	1	5	0
STR13-45	32.5	40	20	23	9	2	6	0
STR13-47	42.5	37	22	26	8	2	5	0
STR13-49	52.5	38	18	31	9	3	1	0
STR13-51	62.5	46	15	21	16	2	0	0
STR13-53	72.5	48	15	18	17	2	0	0
STR13-55	82.5	52	16	17	11	4	0	0
STR13-57	92.5	45	16	22	15	2	0	0
STR13-59	102.5	35	22	22	20	1	0	0
STR13-61	112.5	34	20	26	19	0	0	1
STR13-41B	117.5	37	18	29	14	2	0	0
STR13-42B	135	40	22	22	13	2	0	1
STR13-43B	145	33	19	30	14	2	0	2
STR13-45B	165	37	22	28	11	2	0	0
STR13-47B	185	32	20	29	17	2	0	0

Table 1

Sample	Depth (cm)	Type	Bulk density (g/cm <sup>3</sup> )	SiO <sub>2</sub> (%)	Al <sub>2</sub> O <sub>3</sub> (%)	CaO (%)	Na <sub>2</sub> O (%)	K <sub>2</sub> O (%)	MgO (%)	Fe <sub>2</sub> O <sub>3</sub> (%)	MnO (%)	TiO <sub>2</sub> (%)	P <sub>2</sub> O <sub>5</sub> (%)	LOI (1000 °C) (%)	Ta (ppm)	Zr (ppm)
STR13-41	12.5	Soil	1.0	73.20	12.05	0.16	1.70	4.55	0.21	1.30	0.011	0.18	0.27	4.79	2.3	81.0
STR13-43	22.5	Soil	1.5	69.30	13.49	0.18	2.10	4.96	0.26	1.71	0.021	0.21	0.48	5.51	2.7	73.0
STR13-45	32.5	Soil	1.2	68.50	14.03	0.27	2.40	4.99	0.32	1.69	0.027	0.22	0.59	4.85	2.9	70.0
STR13-47	42.5	Soil	1.7	68.10	14.72	0.33	2.66	5.19	0.36	1.70	0.034	0.22	0.56	4.08	2.4	71.0
STR13-49	52.5	Soil	1.7	70.20	13.99	0.29	2.45	5.09	0.30	1.53	0.032	0.20	0.50	3.22	2.7	67.0
STR13-51	62.5	Saprolite	2.0	70.10	14.36	0.34	2.57	5.05	0.34	1.57	0.032	0.20	0.51	2.96	2.7	62.0
STR13-53	72.5	Saprolite	2.1	70.60	14.08	0.23	2.25	5.30	0.26	1.59	0.030	0.18	0.41	2.52	2.7	62.0
STR13-53 <sup>2</sup>	72.5	Saprolite	2.1	71.20	13.94	0.32	2.39	5.07	0.34	1.60	0.030	0.19	0.42	-	-	-
STR13-53 <sup>3</sup>	72.5	Saprolite	2.1	70.70	13.77	0.34	2.47	5.09	0.34	1.58	0.030	0.18	0.42	-	-	-
STR13-55	82.5	Saprolite	2.1	69.50	14.76	0.29	2.30	5.13	0.39	2.04	0.037	0.22	0.42	2.82	3.3	67.0
STR13-57	92.5	Saprolite	2.4	69.60	14.56	0.32	2.45	5.18	0.39	2.08	0.028	0.20	0.35	2.39	3.1	61.0
STR13-59	102.5	Bedrock	2.5	70.20	14.87	0.35	2.58	5.23	0.20	1.82	0.023	0.20	0.35	2.26	3.1	64.0
STR13-61	112.5	Bedrock	2.6	71.80	14.50	0.30	2.20	5.34	0.20	1.38	0.020	0.16	0.39	2.17	2.8	67.0
STR13-41B	117.5	Bedrock	2.7	71.60	14.80	0.30	2.50	5.35	0.20	1.19	0.020	0.15	0.34	1.97	2.5	64.0
STR13-42B	135	Bedrock	2.7	72.60	14.30	0.30	2.30	5.23	0.25	1.31	0.022	0.18	0.31	1.87	2.6	67.0
STR13-43B	145	Bedrock	2.7	72.60	14.38	0.28	2.37	5.10	0.26	1.25	0.027	0.17	0.33	1.59	2.8	70.0
STR13-45B	165	Bedrock	2.7	72.10	14.19	0.26	2.22	5.16	0.20	1.41	0.032	0.16	0.31	1.83	2.6	77.0
STR13-46B	175	Bedrock	2.7	71.80	14.20	0.30	2.60	5.46	0.26	1.27	0.017	0.15	0.33	1.7	2.5	70.0
STR13-47B	185	Bedrock	2.7	72.40	14.38	0.28	2.33	5.13	0.39	1.16	0.016	0.16	0.32	1.57	2.7	69.0

Table 2

Sample	Depth (cm)	U (ppm)	Th (ppm)	( <sup>234</sup> U/ <sup>238</sup> U)	( <sup>230</sup> Th/ <sup>232</sup> Th)	( <sup>230</sup> Th/ <sup>234</sup> U)	( <sup>226</sup> Ra/ <sup>230</sup> Th)
STR13-41	12.5	3.417	7.800	1.021	1.412	1.036	0.986
STR13-43	22.5	3.621	8.904	1.026	1.433	1.130	1.027
STR13-45	32.5	4.224	9.752	1.025	1.495	1.105	0.928
STR13-47	42.5	3.830	9.636	1.029	1.440	1.158	0.974
STR13-49	52.5	3.763	9.179	1.024	1.471	1.150	0.992
STR13-53	72.5	3.528	8.622	1.034	1.486	1.156	0.931
STR13-53 <sup>2</sup>	72.5	3.575	8.643	1.032	1.499	1.155	0.936
STR13-53 <sup>3</sup>	72.5	3.462	8.699	1.034	1.454	1.162	0.936
STR13-57	92.5	3.841	9.387	1.040	1.519	1.175	0.944
STR13-59	102.5	4.155	9.810	1.036	1.544	1.158	0.949
STR13-61	112.5	3.884	9.111	1.025	1.517	1.140	0.966
STR13-41B	117.5	3.531	8.753	1.016	1.392	1.114	0.999
STR13-42B	135	3.957	8.433	1.024	1.623	1.109	0.963
STR13-43B	145	4.294	9.659	1.015	1.504	1.094	0.963
STR13-45B	165	4.601	9.193	1.011	1.653	1.072	0.984
STR13-46B	175	4.470	9.415	1.006	1.531	1.052	1.008
STR13-47B	185	4.269	9.149	0.987	1.451	1.034	0.988

Table 3

Sample	Depth (cm)	Integrated density (g/cm <sup>3</sup> )	Mass of quartz (g)	Total <sup>9</sup> Be (mg)	<sup>10</sup> Be/ <sup>9</sup> Be	± (%)	In situ <sup>10</sup> Be (at/g quartz)	± (%)
STR11-3	7	0.88	25.02	0.477	1.47E-13	3.1	188144	3.5
STR13-43	22.5	1.19	17.23	0.285	1.55E-13	3.4	171954	3.9
STR13-45	32.5	1.20	19.72	0.293	1.65E-13	4.0	163601	4.5
STR13-47	42.5	1.27	14.68	0.290	1.16E-13	3.9	153162	4.4
STR13-49	52.5	1.34	22.54	0.287	1.70E-13	3.9	144995	4.4
STR13-51	62.5	1.41	18.60	0.277	1.32E-13	3.3	131079	3.9
STR13-53	72.5	1.49	23.79	0.281	1.45E-13	3.8	114030	4.3
STR13-55	82.5	1.56	22.03	0.252	1.23E-13	3.2	97672	3.7
STR13-59	102.5	1.73	22.45	0.303	8.75E-14	3.9	78918	4.4
STR13-61	112.5	1.80	18.32	0.274	6.39E-14	4.0	63992	4.5
STR13-42B	135	1.96	29.31	0.291	8.49E-14	3.9	56360	4.4
STR13-43B	145	2.01	40.35	0.248	1.09E-13	5.2	44712	5.5
STR13-45B	165	2.09	38.00	0.278	7.48E-14	4.5	36532	4.9
STR13-47B	185	2.16	48.84	0.268	8.31E-14	3.9	30440	4.4
STR13-49B	205	2.21	57.09	0.280	8.55E-14	3.8	28057	4.3

Table 4

Site	Regolith production from U-series data (T/km <sup>2</sup> /yr)	Regolith denudation from profile <sup>10</sup> Be data (T/km <sup>2</sup> /yr)	Regolith denudation from river sediment <sup>10</sup> Be data (T/km <sup>2</sup> /yr)	Erosion from river sediment load (T/km <sup>2</sup> /yr)
Strengbach Catchment (France)	30 ± 10 <sup>(1)</sup>	37 ± 15 <sup>(1)</sup>	-	10 ± 2 <sup>(2)</sup>
Susquehanna Shale Hills Observatory (United States)	29 ± 24 - 77 ± 20 <sup>(3)</sup>	27 ± 10 - 34 ± 10 <sup>(4)</sup>	39 <sup>(5)</sup>	-
Betic Range (Spain)	12 ± 2 - 41 ± 9 <sup>(6)</sup>	24 ± 5 - 39 ± 3 <sup>(6)</sup>	-	-
Nunnock Catchment (Australia)	20 ± 3 - 130 ± 24 <sup>(7)</sup>	12 ± 2 - 90 ± 5 <sup>(8)</sup>	87 ± 9 <sup>(8)</sup>	-
Rio Icacos Watershed (Porto Rico)	76 ± 20 <sup>(9)</sup>	42 ± 15 - 85 ± 30 <sup>(10)</sup>	73 ± 25 <sup>(10)</sup>	300 <sup>(11)</sup>

Table 5

## **Mineralogical composition of the analyzed samples : methods and results**

The mineralogical compositions of all the powdered 250- $\mu\text{m}$  bulk samples were determined by XRD with a Siemens D5000 diffractometer with scanning from 4 to 84°, a space step of 0.04° and a time step of 18 s. The <2- $\mu\text{m}$  clay fractions were separated by centrifugation and analyzed after air drying and treatment with glycol ethylene and hydrazine to distinguish illites, smectites, kaolinites and inter-stratified clays. The quantitative estimation of the proportion of the different minerals has been performed by the Rietveld method with the Siroquant software. The analytical uncertainty is approximately 3 % for the mass fraction of quartz and plagioclase and 5 % for the mass fraction of micas and clays.

The whole rock XRD analyses (Table Table EA1) show that quartz is the most abundant primary mineral (32 to 48 %), followed by K-feldspar (17 to 31 %), plagioclase feldspar (15 to 22 %), biotite and muscovite (approximately 9 %). Clay minerals include illites, smectites, interstratified illites/smectites and kaolinites, with a bulk clay fraction from a few % in the bedrock to 10 % in the upper soil (Table 1). Thin section observations confirm the presence of the above primary minerals (quartz, K-feldspar, plagioclase feldspar, biotite, and muscovite) and also indicate the presence of apatite, zircon, small amounts of aluminum silicates, hematite and rutile.

### ***Table Caption***

*Table EA1 1: Bulk X-ray diffraction analysis for the studied weathering profile.*