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Determination of regolith production rates from²³⁸U-²³⁴U-²³⁰Th disequilibrium in deep weathering profiles (Longnan, SE China). Guo-Dong Jia^{1,2}, François Chabaux^{2,1}, Jérôme van der Woerd³, Eric Pelt², Raphaël di Chiara², Julien Ackerer², Zhi-Qi Zhao⁴, Ye Yang¹, Sheng Xu¹, Cong-Qiang Liu¹ 1- Institute of Surface-Earth System Science, Tianjin University, Tianjin 300072, China 2- Laboratoire d'Hydrologie et de Géochimie de Strasbourg (LHyGeS), Université de Strasbourg, CNRS, ENGEES, 1 rue Blessig, 67084 Strasbourg Cedex, France 3- Institut de Physique du Globe de Strasbourg (IPGS), UMR 7516, CNRS, Université de Strasbourg, 5 rue René Descartes, 67084 Strasbourg Cedex, France 4- School of Earth Science and Resources, Chang'an University, Xi'an 710054, China Abstract: 390 words Text: \approx 7630 words

31 Abstract

The present study seeks to evaluate the application of the ²³⁸U-²³⁴U-²³⁰Th radioactive 32 disequilibrium methodology for the determination of the regolith production rates in 33 thick weathering profiles marked by long histories, encountered under various climate 34 regimes, but still very little studied by these techniques. For this purpose, ²³⁸U-²³⁴U-35 230 Th disequilibria have been analyzed in the top 11 m of a lateritic profile developed 36 on a granitic bedrock in south China (Longnan, Jiangxi Province) under a subtropical 37 38 climate. The results demonstrate that in such a weathering profile the determination of weathering rates from the analysis of U-series nuclides in bulk rock samples cannot be 39 recovered by applying in one step to the entire alteration profile the modeling approach 40 classically used to interpret the U-series nuclides, i.e. the "gain and loss" model. The 41 modeling has to be made on subsections of relatively small size (<1 or 2 meters of 42 thickness), so that the model assumptions can be met, especially the constancy of the 43 mobility parameters along the weathering zone. The results also confirm that the upper 44 part of the weathering profiles marked by the vegetation/biological influences and 45 46 responding to the short-term climate variations is not well adapted for applying the Useries nuclides methodology for recovering regolith production rates. Based on the data, 47 regolith production rates were estimated independently on four different deeper zones 48 of the profile. Similar values of ~2m/Ma have been obtained whatever the level, 49 suggesting that such a profile of more than 5 million years would be formed at a 50 relatively stable long-term production rate (averaged over several thousand years). This 51 production rate is slow compared to the production rate deduced from previous in situ 52 ¹⁰Be data, which can be explained by assuming non steady-state erosion of the upper 53 part of the profile. Slow denudation rates similar to the U-series derived production 54 rates of 2 m/Ma can thus be obtained with a minimum exposure time of 60 ky, and an 55 inherited component of 20000 atoms/g originating from the exhumed deeper part of the 56 profile. Altogether the data demonstrate that the combined analysis of U-series and 57 cosmogenic nuclides, which has the potential to become a relevant approach to 58 59 constrain the dynamics of continental surfaces, requires (a) dense and deep sampling for both nuclides studies, and (b) also to consider more systematically the polyphased 60

and variable history of erosion of the continental surface during the Quaternary.

65 1. Introduction

Regolith production and denudation rates are key parameters controlling the response 66 of continental surfaces to environmental modifications, be they anthropic, climatic or 67 tectonic in origin. The knowledge of such time dependent parameters is thus at the heart 68 of numerous present-day scientific issues, for understanding the links between chemical 69 alteration and mechanical erosion processes (e.g., Dixon and von Blanckenburg, 2012), 70 for deciphering the coupling between tectonic exhumation and surface processes of 71 72 alteration and erosion (e.g., Anderson and Anderson, 2010; Owens and Slaymaker, 2014), or for assessing the role of rock alteration in the carbon cycle and thus in the 73 evolution of earth climate (e.g., Berner and Berner, 1996; Berner and Raiswell, 1983). 74 Addressing these scientific issues rest, partly at least, in our capability to develop 75 geochronological tools for constraining time scales of regolith production and 76 denudation. 77

Major improvements were achieved since the middle of the 80s with the 78 development of terrestrial in-situ cosmonuclide analysis, linked to analytical advances 79 80 brought by accelerator mass spectrometry (for more details see the review by Granger et al., 2013 and references therein). These tools became in about 30 years major 81 chronometers in geomorphology to quantify denudation rates on the millennial time 82 scale. The application of these techniques enabled real advances in the understanding 83 of the relations between mechanical erosion and chemical alteration of rocks (Dixon 84 and von Blanckenburg, 2012; Riebe et al., 2017). However, interpretations and 85 conclusions often rely on the hypothesis of a steady state functioning of regolith and 86 alteration profile over the millennial time scale. This remains difficult to test without 87 88 an independent determination of the regolith production rates on similar time scales.

This is possible with the analysis of U-series nuclides, usually ²³⁸U-²³⁴U-²³⁰Th, sometimes complemented by ²²⁶Ra (e.g., Chabaux et al., 2013 and references therein). This type of analysis was developed from the 1990s, in connection with the development of the precise analyses of the U-series nuclides concentrations by thermoionization mass spectrometry and MC-ICP-MS. U-series nuclide based methods enable to determine the alteration time constants at catchment scale with the analysis of river

95 waters and sediments (Dosseto et al., 2006a,b; 2008a; Granet et al., 2007; 2010; Vigier et al., 2001; 2006) as well as at the scale of weathering profiles by the analysis of U-96 series nuclide concentration variations (Chabaux et al., 2003a,b; 2013; Dequincey et al., 97 2002; Dosseto et al., 2008b; Ma et al., 2013). The latter approach has been applied to 98 various alteration contexts: centimetric weathering rinds developed on basaltic clasts 99 (Engel et al., 2016; Ma et al., 2012; 2019; Pelt et al., 2008), metric size spheric alteration 100 systems (Chabaux et al., 2013; Claude et al., 2016), decimetric to metric alteration 101 102 profiles (Ackerer et al., 2016; Dosseto et al., 2008a; Ma et al., 2013; Schoonejans et al., 2016; Rihs et al., 2020). These works have led to the definition of a theoretical and 103 conceptual frame for the application of the radioactive disequilibrium methodology for 104 alteration profiles, which comprises (a) the analysis of the variations of U-series 105 nuclides along the main weathering direction and (b) the interpretation of these 106 variations using a realistic model of radionuclide mobility during water-rock 107 interactions, commonly, a model assuming both gain and loss of elements along the 108 profile (e.g., Chabaux et al., 2008, 2011, 2013; Dosseto et al., 2008b, 2012, 2019; Ma 109 110 et al., 2013). A key assumption behind this approach is to be able to define, within the studied profile or the alteration system, a sufficiently extended zone for which the 111 mobility parameters may be assumed constant. Usually, the analysis of a few samples 112 (3 to 6) in this zone is enough to estimate the nuclide mobility parameters and the 113 sample ages relative to a reference sample (i.e., the deepest one in the profile zone 114 analyzed). The ratio between the thickness and the age of the considered regolith zone 115 gives a mean propagation rate estimate of the alteration front and/or the production rate 116 within the profile, for millennial time scales, which are, as for the cosmonuclides, the 117 118 time scales accessible by the method.

The recent study performed on the Strengbach catchment site (Ackerer et al., 2016), has clearly highlighted the benefit to combine on the same profile both the insitu cosmonuclide analysis (in this case in situ ¹⁰Be in quartz) and the radioactive disequilibria on whole rock. Rates of denudation and regolith production were determined independently establishing firmly the long-term stability of the alteration system. This study demonstrates the importance to carry on these studies using radioactive disequilibria as tools to determine regolith production rates, complementary
and in parallel to the studies using cosmonuclides. This kind of multidisciplinary
approach, combining radioactive disequilibria and cosmonuclides, is certainly one of
the present scientific frontiers to explore by the surface science community if one aims
at crossing new steps towards understanding the rules that govern continental surfaces
functioning (e.g., Riebe et al., 2017).

Such an objective implies to be able to evaluate the applicability of the radioactive 131 132 disequilibria on thick weathering profiles, not well studied so far, although they are present in a wide range of various climatic settings (tropical equatorial to temperate). 133 As recalled above, the studies of the last 15-20 years have mostly dealt with relatively 134 vertically restricted systems of a few meters maximum. Two exceptions however: the 135 first one is the study on the Kaya lateritic profile, Burkina Fasso (Dequincey et al., 2002; 136 Chabaux et al., 2003b) leading to propose an interpretation scheme of radioactive 137 disequilibria in alteration profiles, which makes the basis for following studies. The 138 ²³⁸U-²³⁴U-²³⁰Th disequilibria analyzed in this profile cannot be used in this case to 139 140 determine the regolith production rate but give information on the U redistribution from the surface to the deeper levels, linked to the upper iron cap destabilization since at least 141 800 ka (Chabaux et al., 2003a,b). The second one is the study by Dosseto et al. (2012), 142 achieved on a 15 m-deep profile developed on volcanoclastic rocks in Puerto Rico. 143 Unlike the profile of Dequincey et al. (2002), this profile is not a degrading "fossil" 144 profile. The radioactive disequilibria application leads to the estimates of alteration 145 rates, which are larger than rates obtained on a nearby granodiorite profile (Chabaux et 146 al., 2013). These results and their interpretation rest on the strong assumption that the 147 148 mobility parameters of U and Th are uniform across the whole profile.

The mobility of an element such as U, depends on numerous factors, for instance, the nature of the mineral phases present, the degree of oxidation-reduction of the medium, its pH, the presence of organic and mineral phases that can complex or fix Useries nuclides (e.g., Chabaux et al., 2003a). The diversity of these factors and their probable variation along the weathering profile can obviously make the hypothesis of a constancy of the mobility parameters required by the model difficult to justify over

an entire several meters thick profile. The study of a granodioritic profile in Puerto Rico 155 (Chabaux et al., 2013) illustrates this point. The profile is marked by a spheroidal 156 alteration, with two contrasted alteration zones: a zone in contact with the bedrock, 157 made of about 20 layers of a few cm each and a 1 to 2 m-thick zone of unconsolidated 158 laterite, developed towards the surface from these layers. The authors show that the 159 gain and loss model can be applied on each of these two zones, but with different 160 mobility parameters for each of them. Based on this study, along with a few following 161 ones (i.e, Schoonejans et al., 2016; Ackerer et al., 2016; Gonthier et al., 2015), we 162 propose that for several meters thick profiles, often marked by significant mineralogical 163 changes related to the differential weathering of minerals, it is necessary to apply the 164 U-series methodology not to the profile as a whole with a limited number of samples, 165 but on sub-zones of relatively limited thickness within the profile, each ones with a 166 sufficiently dense sampling (3 to 6 samples). We also anticipate that such an approach 167 conducted on different sub-zones of a single weathering profile can be a relevant 168 method for assessing its long-term stability, as the study on the Puerto Rico profile also 169 170 suggests (Chabaux et al., 2013).

These are the working assumptions that are tested in the present study by analyzing the upper 11 m of a lateritic profile developed on a granitic bedrock in south China, under a subtropical climate, and already well studied and characterized in term of mineralogy and geochemistry (Cui et al., 2016; Liu et al., 2016; Zhang et al., 2015). In addition, we will compare our results of regolith production rates with the already published denudation rate constrained by in situ ¹⁰Be cosmonuclide for this profile (Cui et al., 2016) and discuss the meaning of the observed differences.

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179 **2. Study area and sample selection**

The JLN-S1 profile targeted in this study has been previously analyzed by Zhang et al. (2015), Cui et al. (2016) and Liu et al. (2016). It is located in the northern Nanling granite belt in Longnan, Jiangxi Province, southeastern China (114°46'E, 24°58'N), at an altitude of 225 m asl (Fig. 1). The study area belongs to a hilly area of moderate relief (altitudes from 600 to 800 m; highest peak at 1200 m asl 50 km to the south at

the sources of the Tao Jiang, a tributary of the Yangtse) in a subtropical zone marked 185 by a humid summer monsoon climate and mostly covered by a subtropical evergreen 186 broadleaf forest. At the study site, the mean annual precipitation (MAP) is 1500 mm 187 and the mean annual temperature (MAT) is 19°C, with monthly temperature ranging 188 from 8.3°C in January to 27.7°C in July (e.g., Schamm et al., 2014). The profile is 189 developed on a K-feldspar granite, formed during the Yanshanian orogeny (end of early 190 Jurassic) and dated at 178.15 ± 0.84 Ma (Rb-Sr isochron age; Fan and Chen, 2000). The 191 192 granite is a medium to coarse-grained granite with porphyritic-like texture. The main phenocrysts are quartz, K-feldspar and plagioclase with small amounts of biotite and 193 hornblende. Magnetite, ilmenite, zircon, fluorite and tourmaline are present as 194 accessory minerals (Chen et al., 2004). 195

196 The JLN-S1 profile is exposed in a road cut above the dammed Tao Jiang riverbed (Fig.1). It is located 150 m away from the summit at the bottom of a ridge with a 197 relatively steep slope (15-18° in Cui et al., 2016). Detailed petrological and 198 mineralogical description can be found in Zhang et al. (2015), Cui et al. (2016) and Liu 199 200 et al. (2016). In short, the JLN-S1 profile corresponds to the top 11 m of a granitic weathering profile. Liu et al. (2016) subdivided the JLN-S1 profile into three main 201 horizons, a saprolite horizon from 11 m to 4 m, a massive soil horizon from 4 m to 1 m 202 and a soil horizon (<1m) (Fig. 1c). A deeper weathered bedrock horizon is not reached 203 at the JLN-S1 outcrop but deduced from bedrock samples collected close to the bottom 204 of the profile in a nearby streambed. The base of the JLN-S1 profile is assumed to be 205 close to the transition with the deeper weathered bedrock horizon (in Liu et al., 2016). 206 The mineralogy and major element variations allowed the authors to delineate the 207 weathering reaction sequence along the profile (Liu et al., 2016). The weathered 208 bedrock is characterized by dissolution of biotite, hornblende, and apatite. The saprolite 209 horizon is marked by plagioclase dissolution mainly from 11 m to ≈ 6 m depth and by 210 kaolinite precipitation all along the horizon. The massive soil horizon is dominated by 211 K-feldspar weathering and also by desilication and conversion of kaolinite into gibbsite. 212 213 The overlying soil is characterized by primarily dissolved quartz and presence of bioturbation. The depth of 1 m is also considered by Liu et al. (2016) as the maximum 214

depth at which lateral movement of materials moving downslope could be detected. In
addition, the authors have noticed the presence of a quartz vein at a depth of 170–190
cm, and of a vein dominated by plagioclase at a depth of around 700 cm.

For the purpose of our study, it is necessary to define different sub-zones in the 218 profile on which applying independently the U-series methodology. The mineralogical, 219 granulometric and geochemical data show a fairly clear boundary at 4 m depth between 220 the saprolite horizon and the massive soil horizon defined in Liu et al. (2016) (Fig. 2). 221 222 In addition, the quartz and plagioclase veins, which crosscut the profile at 2 m and 7 m depths, respectively, create two mineralogical discontinuities within the profile, which 223 may impact the assumptions of uniformity of mobility parameters that the model 224 requires. For these reasons, we have subdivided the saprolite into two distinct zones the 225 zone above and below the limit at 7 m depth. For similar reasons, we have subdivided 226 the upper zone into two subzones, the one below the quartz vein (between 4 m and 2 m) 227 and the one above the quartz vein (<1.7 m), eventhough it has been shown that the soil 228 horizons are not the best ones to apply the U-series approach (Ackerer et al., 2016; 229 230 Gontier et al., 2015). We have therefore defined four *a priori* zones in the profile for applying the U-series methods. They are reported in Fig. 2. In each of these zones, a 231 minimum of 5-6 samples have been selected. The samples are those initially collected 232 for the studies by Zhang et al. (2015) and Liu et al. (2016) following the sampling 233 strategy published in their papers. Mineralogical composition and major element 234 concentrations of most of these samples are given in the above referenced studies. 235

236

237 3. Analytical and modeling methods

238 **3.1 Uranium and thorium analyses**

Uranium and thorium concentrations and isotope ratios were analyzed at the Laboratoire d'Hydrologie et de Géochimie de Strasbourg (LHyGeS), University of Strasbourg and CNRS, France, following a procedure described in Bosia et al. (2018), adapted from previously published ones (Pelt et al., 2013; Granet et al., 2010; Chabaux et al., 1995, 2019). Approximately 100 mg of finely powdered samples were weighted and spiked with a mixed pure artificial ²³³U-²²⁹Th spike and completely digested in low245 pressure teflon beakers using a multi-step combination of HF, HNO₃, HClO₄, HCl and H₃BO₃ acids at 100-200°C. The 233 U- 229 Th spike was consistently calibrated at a ~5‰ 246 level both via certified NIST gravimetric solutions (SRM3164U and SRM3159Th) and 247 via ATHO rock standard using data from Williams et al. (1992). Uranium and thorium 248 were separated and purified by a multi-step conventional ion exchange chromatography 249 using Biorad AG1x8 200-400 mesh anionic resin. U and Th isotopes were measured on 250 a plasma mass spectrometer (MC-ICP-MS Neptune) by bracketing method respectively 251 against the natural IRMM-184 U and IRMM-035 Th solutions. As in Cogez et al. 252 (2018), the $(^{234}U/^{238}U)$ activity ratio used for the IRMM-184 standard is the mean 253 TIMS-Triton value measured at the LHyGeS (internal error-weighted mean $0.9698 \pm$ 254 0.0011, 2SE, N=8) instead of the last certified value from Richter et al. (2005) (0.9682 255 \pm 0.0006). The latter modification yielded better means for the (²³⁴U/²³⁸U) activity ratio 256 measured in the secular-equilibrium HU1 standard solution that was 1.0010 ± 0.0022 257 (2SD, N=21, 50-70 ppb U) during this study. The IRMM-036 Th standard solution was 258 run 4 to 5 times during each of the 4 Neptune Th-sessions yielding a ²³²Th/²³⁰Th ratio 259 of 326483 ± 1932 (2SD, N=19, 40 ppb Th) consistent with values from Sims et al. 260 (2008) and Carpentier et al. (2016). 261

Powder digestion replicates of samples and BCR-2 rock standard were used to 262 determine the global precision and accuracy of U and Th isotopic measurements. For 263 this study, the average values of U-Th concentrations and $(^{234}U/^{238}U)-(^{230}Th/^{238}U)$ 264 activity ratios for the BCR-2 rock standard are $1.698 \pm 0.008 - 5.865 \pm 0.021$ ppm and 265 $0.995 \pm 0.004 - 1.000 \pm 0.012$ (2SE, N=4), respectively, consistent with previous values 266 from LHyGeS laboratory and from other laboratories (Rihs et al., 2020; Sims et al., 267 2008; Matthews et al., 2010) and consistent within 5‰ with secular equilibrium. 268 Procedural blanks starting just before the digestion step range from 13 to 75 pg and 361 269 to 494 pg for U and Th, respectively, and are negligible compared to the amount of U 270 and Th processed. 271

272

273 **3.2** Determination of weathering rates and regolith production rates from ²³⁸U-

274 ²³⁴U-²³⁰Th disequilibria

For the present study, the determination of the weathering and regolith production rates 275 from the analysis of ²³⁸U-²³⁴U-²³⁰Th disequilibria in weathering profiles is based on the 276 approach classically used for such a purpose, which has been detailed in several 277 previous studies (e.g., Ackerer et al., 2016; Chabaux et al., 2011, 2013; Dosseto et al., 278 2008b; Ma et al., 2012, 2013). In such an approach the time variation of radioactive 279 nuclides for a sample in a weathering zone, is described in terms of continuous 280 processes involving, in addition to the radioactive decay laws, loss and gain processes 281 represented by first-order kinetic rate laws for the loss processes and zero-order kinetic 282 rate laws for the gain processes. The loss processes may include many different 283 processes such as mineral dissolution or nuclide desorption from different minerals, 284 and the gain processes atmospheric dust deposition, illuviation processes, sorption and 285 co-precipitation of nuclides on and within different minerals such as Fe-Mn-oxides, 286 clay minerals for instance. For the modeling approaches all the gain (resp. loss) 287 parameters are lumped into only one bulk parameter. The time variation of the ²³⁸U-288 ²³⁴U-²³⁰Th nuclides concentration is thus described with the three following equations: 289

$$\frac{d^{238}U}{dt} = f_{238}{}^{238}U_0 - k_{238}{}^{238}U - \lambda_{238}{}^{238}U(1)$$
$$\frac{d^{234}U}{dt} = f_{234}{}^{234}U_0 + \lambda_{238}{}^{238}U - k_{234}{}^{234}U - \lambda_{234}{}^{234}U(2)$$
$$\frac{d^{230}Th}{dt} = f_{230}{}^{230}Th_0 + \lambda_{234}{}^{234}U - k_{230}{}^{230}Th - \lambda_{230}{}^{230}Th(3)$$

where λ_i , k_i and f_i are the radioactive decay, loss and gain constants (in yr⁻¹), 290 respectively, for the radionuclides i (i.e., 238 U, 234 U, 230 Th). In the equations, t is the 291 time elapsed between the reference weathering state and its current state. For 292 simplification during the equation-solving procedure, the input fluxes are expressed as 293 a proportion of the number of atoms of nuclides added per year to the initial sample. 294 The loss (k) and gain (f) terms used in the equations are usually assumed to be constant 295 with time (e.g., Chabaux et al., 2013; Dequincey et al., 2002; Dosseto et al., 2008b; Ma 296 et al., 2013). The mean production rate of regolith, P (in mm/kyr), can be estimated 297

through the resolution of the above equation system and the determination of the age Δt (in kyr) of a sample relative to a reference sample from deeper in the profile at a distance Δh (in mm) along the weathering direction. For isovolumetric weathering, P (mm/kyr) is given by:

$$P = \frac{\Delta h}{\Delta t} \ (4)$$

The gain and loss coefficients in equations 1 to 4 are usually not known. The ²³⁸U-²³⁴U-302 ²³⁰Th analyses yield only two independent data per sample, i.e., the (²³⁴U/²³⁸U), and 303 $(^{230}\text{Th}/^{234}\text{U})$ ratios. The analysis of only one sample (in addition to a reference sample) 304 is thus not sufficient to determine the weathering production rate P. The mathematical 305 formalism developed to retrieve such time information assumes that the gain and loss 306 coefficients (k_i and f_i) of the model can be considered constants, at least for a part of the 307 profile from which several different samples can be collected and analyzed. The 308 measured $(^{234}\text{U}/^{238}\text{U})$ and $(^{230}\text{Th}/^{234}\text{U})$ ratios of each sample are used to determine the 309 mobility parameters of the model and the age of different samples relative to the 310 reference sample. The numerical solutions are obtained using a stochastic quantum 311 particle swarm optimization scheme, the principle of which has been detailed in 312 313 Chabaux et al. (2012, 2013) and is summarized in §4-3. As recalled and illustrated by Ackerer et al. (2016), such modeling approaches only allow for determination of mean 314 long-term values, i.e., averaged over several kyr or tens of kyr, for nuclide gain and loss 315 parameters and for regolith production rates. The approach does not assume constant 316 317 weathering rates. The ages of each sample as compared to the reference sample are determined independently. 318

319

320 **4. Results**

Uranium and thorium concentrations and $(^{234}U/^{238}U)$, $(^{230}Th/^{234}U)$ and $(^{230}Th/^{232}Th)$ activity ratios in the regolith and soil samples are given in Table 1.

323

324 4.1 U, Th concentrations and activity ratios

325 At the profile scale, uranium and thorium concentrations range from 3.2 to 8.6 ppm and

27.4 to 53.8 ppm, respectively. Variations in Th concentrations along the profile do not 326 show any significant increasing or decreasing trend with depth except perhaps in the 327 upper meter of soil. More importantly, the Th/Ti ratio is almost not variable in this 328 profile, without showing any systematic variation along the profile, even across each of 329 the different zones. As Ti is considered as an immobile element regarding weathering 330 in the JLN-S1 profile (Liu et al., 2016; Zhang et al., 2015), the fairly constancy of the 331 Th/Ti ratio along the profile indicates a relative immobility of Th in the upper 11 m of 332 333 this profile (Fig.3c). The U/Th ratios show a higher variability along the profile with the U/Th ratio ranging from 0.1 to 0.2. 334

It can be observed a slight but systematic decrease of the U/Th ratio from ~400 cm 335 to the surface, which could indicate U leaching processes affecting the profile, at least 336 in its upper part (Fig. 3d). In the deepest horizons, the U/Th ratio is more variable, 337 especially between 7 m and 4 m depth with significant variations over short distances. 338 Without a more detailed mineralogical knowledge of the profile, which is beyond the 339 scope of the study, it is difficult to constrain the origin of such variations. They could 340 341 indicate differences in some of the mineralogical characteristics of the parent rocks at the origin of these horizons, relative to the other horizons. They could also indicate 342 different and variable U mobilities over short distances in these intermediate and deeper 343 levels. These characteristics are important to keep in mind when discussing the 344 applicability of our approach to these intermediate levels of the profile. 345

 $(^{234}U/^{238}U)$ and $(^{230}Th/^{234}U)$ activity ratios range from 0.936 to 1.014 and from 346 0.814 to 1.122, respectively (Table 1, Fig. 4). In the deeper part of the profile (deeper 347 than ~800 cm) these ratios have rather constant values scattered around 0.94 for 348 (²³⁴U/²³⁸U) and 1.07 for (²³⁰Th/²³⁴U). Above, the U activity ratio increases to secular 349 equilibrium ($(^{234}U/^{238}U) = 1$) at around 600 cm depth, then remains close to the 350 equilibrium value. Above 135 cm, $(^{234}U/^{238}U)$ is > 1 and variations are small. For 351 $(^{230}\text{Th}/^{234}\text{U})$, between 800 cm and the surface, three zones can be defined. From 800 to 352 400 cm, $(^{230}\text{Th}/^{234}\text{U})$ decreases from 1.12 to 0.93; then increases to reach 1 at ≈ 150 cm 353 depth; in the uppermost horizons (>135 cm), both $(^{230}\text{Th}/^{234}\text{U})$ and $(^{234}\text{U}/^{238}\text{U})$ ratios 354 are relatively constant with values greater than 1. The depth variation of $(^{230}\text{Th}/^{232}\text{Th})$ 355

(Fig. 4) looks like that of U/Th with relatively limited variations (0.33 to \approx 0.4) in the deeper (1100 to 700 cm) and upper part (>400 cm) of the profile and a much larger scatter, in the intermediate part of the profile between 700 and 400 cm depth, where (²³⁰Th/²³²Th) ranges from 0.35 to \approx 0.54.

360

361 4.2 Modeling results

As explained in section 2 four a priori zones have been defined in the profile for 362 interpreting the variations of the ²³⁸U-²³⁴U-²³⁰Th disequilibria within each of these 363 zones using the modeling approach detailed in section 3.2. The small variations of the 364 Th/Ti ratios along the profile suggest a low mobility of Th during weathering processes 365 within the profile. For all the simulations, Th has been therefore considered as immobile, 366 which imposes $f_{230} = k_{230} = 0$ in the modeling equations. The initial (or a priori) variation 367 ranges for the other model parameters are 10^3 to 10^6 yr for final age, 10^{-9} to 10^{-3} yr⁻¹ for 368 k_{238} and k_{234} , and 10^{-12} to 10^{-5} yr⁻¹ for f_{238} and f_{234} . In addition, an a priori variation range 369 from 0.5 to 3 has been retained for the two k_{234}/k_{238} and f_{234}/f_{238} ratios. 370

371 Using such initial constraints, application of the modeling approach on each of the four initially defined zones shows that only in zone 2 the $(^{234}U/^{238}U)$ and $(^{230}Th/^{234}U)$ 372 activity variations can be simulated correctly with the same set of parameters for the 373 whole zone (Fig. 5). For the two deeper ones, this was not possible. In the deepest zone, 374 simulations are only feasible by dividing this zone into two new sub-zones (4a-4b) (Fig. 375 5). The variations of the ²³⁸U-²³⁴U-²³⁰Th disequilibria within zone 3 are also impossible 376 to simulate with a single set of parameters all along the zone. For this part of the profile, 377 the model as described by equation 1 can be applied on only a limited thickness of zone 378 3, i.e., on the upper part of the plagioclase vein and slightly above (sub-zone 3a, Fig. 379 5). The uppermost zone of the profile (zone 1 in Fig. 2) is marked by very small 380 variations of (²³⁴U/²³⁸U) and (²³⁰Th/²³⁴U) activity ratios, which prevent any precise 381 simulation. This zone will thus no longer be considered in the following. We will thus 382 model only zone 2 and the 3 sub-zones 3a, 4a and 4b (Fig. 5). 383

For each of these zones the modelling approach has been applied following the procedure described in Chabaux et al. (2013). The unknown parameters, i.e., the k and

f mobility parameters of 238 U and 234 U nuclides, and the age of the different samples 386 relative to the reference sample, are determined by a Quantum-behaved Particle Swarm 387 Optimization (QPSO) algorithm (e.g., Sun et al., 2004). It is a population-based 388 evolutionary technique in which each particle or bee is provided with a set of 389 parameters. The technique does not work on individual particles only, but on a set of 390 particles (named swarm) moving in a multidimensional search space. At each time step, 391 the whole swarm is tested against the model. Each particle is used to calculate 392 393 theoretical activity ratios from the equations 1 to 4 and the theoretical values are compared to the measured ones at the least squares sense, i.e., by calculating the 394 objective function defined as the sum of the squares of the residues between measured 395 and calculated values. The retained simulations are those whose objective function is 396 below a predefined threshold value. In the developed approach, to provide a more 397 comprehensive learning algorithm, particles are allowed to share memories of their best 398 positions in a local neighbourhood. Therefore, an optimal solution emerges from the 399 swarm. 400

401 For the first step of the simulation, particles are randomly distributed in the a-priori parameter space, defined from the a priori variation ranges retained for the different 402 model parameters (see above). In order to respect a satisfactory balance between 403 exploration (local minima) and exploitation (best fit) of the solution space, we 404 performed for each modelled zone 10 to 20 independent simulations, leading to 10 to 405 20 independent minimizations and we retained for each independent simulated swarm 406 the 5000 solutions that fit data under a recruitment criterion initially fixed at 3% for the 407 objective function. 50,000 to 100,000 admissible solutions are therefore recovered for 408 each modelled zone of the profile. The criterion of 3% was defined to ensure a satisfying 409 fit with respect to error bars. We have however verified that the results of the 410 simulations do not differ whether we use a threshold of 3% or 1.5% for the objective 411 function. This indicates the globally convergent behaviour of the method. 412

In order to estimate means and standard deviations of simulated values that are statistically meaningful a Kernel Density Estimation (KDE) fitted on a normal distribution was applied to the entire population of the 50,000 (100,000) solutions

recovered by the simulation procedure. The values for the production rate, the different
mobility parameters as well as of some of their ratios are given in Table 2 for the 4
modelled zones.

The results show that the weathering production rates are of similar magnitude for 419 the 4 modeled zones (Table 2), with values of ~ 2 m/Ma (1.85±0.59 to 2.66±0.23 m/Ma). 420 The mean values of gain (f) and loss (k) parameters derived from the simulation for U 421 range from 9.504×10^{-7} to 5.249×10^{-6} y⁻¹ for k_{234} , from 5.645×10^{-7} to 3.520×10^{-6} y⁻¹ for 422 k_{238} , from 8.929×10⁻⁷ to 2.522×10⁻⁶ v⁻¹ for f_{234} and from 6.045×10⁻⁷ to 2.090×10⁻⁶ v⁻¹ 423 for f_{238} . They fall into the range of previously published values for shale, granodiorite 424 and granite (Ackerer et al., 2016; Chabaux et al., 2013; Dequincey et al., 2002; Dosseto 425 et al., 2008b; Ma et al., 2013). The values tend to vary along the profile with loss and 426 427 gain parameters higher and lower, respectively, in the deeper zones than in the upper ones (Table 2). The difference of the mobility coefficients between the zones are, 428 however, much more apparent when compared to the k_{238}/f_{238} and k_{234}/f_{234} ratios (Fig 6), 429 with values systematically higher than 1 in the deeper parts of the profiles (zones 4a 430 431 and 4b) and lower than 1 in the upper parts (zones 2 et 3a).

432

433 **5. Discussion**

434 **5.1 Application of the modeling approach**

The results obtained for this study clearly validate our working hypothesis that in such 435 relatively thick weathering profiles the determination of weathering rates from the 436 analysis of U-series nuclides in bulk rock samples cannot be recovered by applying the 437 modeling approach in one step to the entire alteration profile. The modeling has to be 438 made on subsections of relatively small size, so that the model assumptions can be met, 439 especially those concerning the constancy of the mobility parameters along the 440 weathering zone. As highlighted with the above results, the mobility parameters vary 441 along the profile, with significant differences in the massive soil zone (zone 2) and the 442 deep saprolite zone (zones 4a, 4b) for both U gain and loss parameters, which result in 443 444 systematic differences in the k/f ratios between the upper and deeper zones (Fig. 6). Liu et al. (2016) have characterized the sequence of weathering and transformation of main 445

major minerals in the JLN-S1 profile. The resulting differential weathering sequence 446 lead to mineralogical zonation along the profile, which might be invoked to partly 447 explain the observed mobility parameter variations. Thus, the higher U loss coefficients 448 in zones 4a and 4b than in the massive soil (zone 2) and in zone 3a, could result from a 449 relatively larger dissolution of primary minerals in the JLN-S1 deep level (Liu et al., 450 2016), compared to the upper levels, as illustrated for example by the plagioclase 451 variation curve given in Fig. 2. Similarly, the larger U gain parameters in the shallower 452 453 zones 2 and 3a than in the deeper ones, might explain the gibbsite precipitation in the upper part of the profile (Liu et al., 2016), which can significantly adsorb U. 454

The modeling results support also the conclusions of previous works (Ackerer et 455 al., 2016; Gontier et al., 2015; Rihs et al., 2016) that the upper part of the weathering 456 profiles marked by the vegetation/biological influences is not well adapted for applying 457 the U-series nuclides methodology for recovering regolith production rates. More 458 importantly, they also indicate that below the vegetation influence zone, the zones 459 suitable for application of the modeling approach do not exceed 1 or 2 meters in 460 461 thickness, and that all parts of the profile are not necessarily appropriate for such modeling. This is certainly related to the spatial variability of the parameters controlling 462 the mobility of an element such as U in a weathering profile. In addition to the physical 463 and chemical parameters of the medium (pH, Eh), which can indeed vary over short 464 distances, metric or less, the nature of the mineral phases present and their spatial 465 variability are also important to consider. In a weathering profile, these mineral phases 466 control the capacity of U to be released by dissolution of the U-carrier phases or to be 467 fixed on secondary mineral phases. In the JLN-S1 profile, the differential weathering 468 469 processes as well as the initial lithological heterogeneity, which both may cause mineral paragenesis variations over quite short distances, may be invoked to explain the low 470 vertical extension of the zones to which the proposed model for interpreting U-Th 471 disequilibria variations can be applied. For instance, in zone 4, the deepest part (4b) is 472 marked by a rather significant variation of plagioclase proportions with depth, while in 473 474 the overlying zone (4a) the plagioclase content is fairly constant. Such differences certainly indicate weathering processes or even differential weathering sequences, 475

significantly different between these two zones, which can easily explain why 476 simulation of ²³⁸U-²³⁴U-²³⁰Th disequilibria variations along zone 4 must be done on the 477 two subzones with two different sets of mobility parameters. In addition, zone 3 is 478 marked by a strong lithological heterogeneity due to the presence of a plagioclase vein. 479 The mineralogical variations that such a vein induce may result in a significant variation 480 of the mobility parameters across zone 3, which is too strong compared to the number 481 of analyzed samples for allowing the application of the modeling approach in this zone, 482 483 outside the subzone 3a. These different results thus clearly evidence that in thick weathering profiles the degree of mineralogical variability along the profile can become 484 a critical not to say a limiting factor for the applicability of the methodology of U-series 485 disequilibrium on bulk rock samples to recover regolith production rates. The above 486 approach at least for profiles developed on granitic bedrock can therefore only be 487 applied to sections of relatively limited vertical extension. As a consequence, the profile 488 parts over which the simulations can be performed, at least in the case of the Longnan 489 profile, are narrow layers where the variations in $(^{234}U/^{238}U)$ and $(^{230}Th/^{234}U)$ ratios are 490 491 relatively small compared to the analytical uncertainties. This explains why a very large number of simulations can fit the data and therefore why the alteration rates and 492 mobility parameters obtained by this approach cover a wide range of possible values. 493 Although more constraints can be considered by perhaps increasing the number of 494 samples per sub-area (e.g., in subzones 4a or 4b) this intrinsic characteristic of the data 495 will not be modified and the application of this approach at least in this study case will 496 mainly give orders of magnitude for the model parameters. Nevertheless, as detailed in 497 the following section, these orders of magnitude are sufficient to discuss the 498 499 relationships between the regolith denudation and production rates.

500

501 **5.2** Comparison of long-term regolith production and denudation rates

As presented in the results section, a weathering front propagation rate can be determined in four different zones (2, 3a, 4a, 4b), leading to the determination of four different rates, but of the same order of magnitude, with values of ~ 2 m/Ma. Such an observation implies that weathering in the JLN-S1 profile propagates at similar rates

506 along the studied zone, i.e., from 10 m depth, bottom of zone 4b to 2 m depth, summit of zone 2, and that a sample moves up from the bottom of this 8 m-thick zone to its 507 summit in about 4 million years. If we admit that this rate is about the same on the 508 whole profile outcrop and by assuming, as explained in Liu et al. (2016), that the 509 bedrock-saprolite transition zone is close to the outcrop base, the 11 m-thick profile 510 may reach a minimum age of 5.5 million years near its top. The feasible simulations on 511 this profile were applied on sections of small vertical extension, usually ≤ 1 m, which 512 513 correspond to durations of about 500 ky, while the sample vertical thickness of about 20 to 50 cm represents an average time window of 100 to 250 ky. Such a loose time 514 resolution, associated with the large uncertainty on the rate determination, makes it 515 difficult to accurately evaluate by this approach the impact of environmental processes 516 varying over shorter time scales, such as the quaternary climate fluctuations (10-20 ka, 517 e.g., Lisiecki and Raymo, 2005), or the anthropogenic activity (<10 ka), on the 518 weathering rates. On the other hand, the similarity of weathering rates obtained in the 519 present study from the bottom of the profile to about 2 m depth implies that the average 520 521 long-term millennial rates did not vary a lot over the last 4 million years, at least in the deeper part of the profile. In other words, this means that on such long-time scales and 522 in its deeper part the weathering profile is close to "steady state". At first order, this is 523 consistent with the fact that the geographical area is tectonically stable since the end of 524 the Paleogene (≈ 23 Ma; e.g., Faure et al., 2017), and that the modern climatic pattern, 525 controlled by the East Asian monsoon, was established since at least the Miocene-526 Pliocene transition 5 Ma ago, with alternations of wet and dry periods during the 527 Pleistocene and a currently subtropical climate since ≈ 10 ka (e.g., Wang et al., 2016). 528

As recalled in §3.2, for an isovolumic weathering at steady-state the weathering rate determined by the radioactive disequilibrium methodology corresponds directly to the regolith formation rate. For the studied profile, the isovolumic character has been validated for most of the first 4 m, notably in the massive soil unit (zone 2 in our study) in Cui et al. (2016). It is thus tempting to extend this result to the rest of the profile and to assume isovolumic weathering in the underlying more compact rocks. With this assumption and on the basis of our estimates, a long-term regolith production rate of

- ≈ 2 m/Ma can thus be proposed for the weathering profile for the last 4-5 million years determined at best with a resolution of a few 10 kyrs.
- Such a result indicates therefore that the regolith production rate calculated in the 538 present study with U-series nuclides on the deeper part of the JLN-S1 profile is about 539 10 times slower than the denudation rate estimated on the upper part of the same profile 540 by Cui et al. (2016) using in-situ produced cosmogenic nuclides. More precisely, the 541 denudation rate was determined from 6 samples collected along the uppermost 4 m of 542 the JLN- S1 profile by interpreting the depth variations of the in situ ¹⁰Be concentrations 543 with the usual depth profile equation (eq. 1 and 2 in Cui et al., 2016) assuming steady-544 state. The authors also considered, as classically done in many previous studies (e.g., 545 Brown et al., 1995; Granger and Smith, 2000; Braucher et al., 2009), that bedrock 546 alteration profiles do not bear inheritance prior exposure. Such assumptions result in a 547 modeled denudation rate of 17.7 ± 0.5 m/Ma and a minimum exposure time of $349 \pm$ 548 26 ka (Fig. 7a; modeling following Yang et al., 2019, 2020), values very similar to 19 549 \pm 3 m/Ma and \approx 300ka in Cui et al. (2016). The JLN-S1 profile would thus be 550 551 characterized by an important discrepancy between the regolith production rate determined on the lower part of the profile and its denudation rate estimated on its upper 552 part. Such differences would no more allow this profile to be considered as being at 553 steady state: the thickness of the weathering profile would decrease with time and the 554 10 m thick profile would disappear in about 600 ky. 555
- However, with a similarly precise fit to the ¹⁰Be data (Fig. 7a), but accounting for 556 inheritance (e.g., Ackerer et al., 2016; Yang et al., 2020), a slower denudation rate 557 similar to the U-series derived production rates of about 2 m/Ma is obtained with a 558 minimum exposure age of 60 ky (best estimate 60-76ka) and considering an inherited 559 concentration of 10 Be of $16-21*10^3$ atoms/g. Such inheritance is in this case not due to 560 prior near surface exposure, but to ¹⁰Be accumulation from muonic production at great 561 depth over long periods. Such a model implies that the profile has suffered stripping of 562 its top layers (about at least 1 m) during a short period ≈ 60 ky ago and that since that 563 time it is characterized by a relatively slow denudation rate (≤ 2 m/Ma). 564
- 565 Based on the above interpretation that reconciles the ²³⁸U-²³⁴U-²³⁰Th disequilibria

and in situ ¹⁰Be data, we propose that the profile has reached a relatively long-term 566 stability, over the last few millions of years, with long term denudation and production 567 rates of $\approx 2m/Ma$, episodically interspersed with fast and short intense denudation 568 periods (Fig. 8). This scenario is similar to the interpretation proposed by Ackerer et al. 569 (2016) from the combined analysis of U-Th-Ra disequilibrium and in situ ¹⁰Be 570 concentration in a 2 m-deep weathering profile sampled at the summit of the granitic 571 Strengbach catchment (Vosges mountain, France). The authors show that the ¹⁰Be 572 concentration variation with depth implies similar denudation and production rates for 573 the regolith, with an exposure age of about 20 ka and an average inheritance in 574 agreement with a short period of erosion of the upper part (2-4 m) of the profile about 575 20 ka ago, linked to the climatic variation in this mountainous region in Europe, in 576 particular the last glacial maximum. The JLN-S1 profile, is located in a tropical climate, 577 where glacial or peri-glacial processes cannot be responsible for the change in erosional 578 processes. We thus make the assumptions that the production and denudation rates of 579 about 2 m/Ma correspond to an average long-term rate, resulting from long phases of 580 581 low erosion, similar to present-day conditions with abundant vegetation cover under a warm and humid climate, and short phases of stronger erosion, when the climate 582 changes are sufficient to significantly impact the density of the vegetation cover, 583 probably during dry periods due to monsoon weakening. The return periods of these 584 intense erosional events might be linked to the intensity of the regional changes of the 585 vegetation cover and/or the regional climatic conditions induced by the global climate 586 cycles of the Quaternary (Fig. 8). 587

Validating this interpretation, however, will certainly need more appropriate 588 sampling of the profile (e.g., Ackerer et al., 2016), namely in its lower part, to better 589 constrain the average inheritance and the ¹⁰Be muonic production at great depth (e.g., 590 Heisinger et al., 2002a,b; Braucher et al., 2013; Lupker et al., 2015), but also in its 591 upper part to better assess the recent erosion and denudation, and the near-surface 592 perturbation of the soil layers as possibly reflected in the ¹⁰Be concentration scatter of 593 the data (see for instance, figure 3 in Cui et al., 2016). This is clearly outside the scope 594 of this study focused on the analysis of U-series nuclides to determine regolith 595

production rates. The important point to be stressed at this stage is (1) that the ¹⁰Be data 596 is not necessarily in contradiction with a long-term steady-state functioning of the 597 studied profile, and (2) that the analysis of U-series disequilibria in the "deep" levels of 598 weathering profiles, i.e. below the horizons strongly influenced by erosion processes, 599 is certainly well adapted to determine long-term regolith production rates. The JLN-S1 600 results also emphasize that the future development of the combined analyses of U-series 601 and cosmogenic nuclides, which are important to constrain the long-term dynamics of 602 continental surfaces, requires dense samplings with depth for both nuclides studies, 603 certainly denser than what has been usually performed to date. They also indicate that 604 the correct interpretation the ¹⁰Be data in such granitic context requires to consider more 605 systematically the importance of muonic accumulation of ¹⁰Be at depth, in relation to 606 the polyphased history of erosion of the continental surfaces during the Quaternary. 607

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609 Conclusions

The analysis of ${}^{238}\text{U}{}^{-234}\text{U}{}^{-230}\text{Th}$ disequilibria in a ≈ 11 m-deep weathering profile 610 611 developed on a granitic bedrock in south China (Longnan, Jiangxi Province) allow us to discuss the application of the radioactive disequilibria methodology for determining 612 regolith production rate to thick weathering profiles. The results clearly evidence that 613 for such deep weathering profiles the determination of regolith production rates from 614 the analysis of U-series nuclides in bulk rock samples cannot be recovered with a single 615 interpretative model of the entire alteration profile by the modeling approach classically 616 used to interpret the U-series nuclides variation along a profile, i.e., the "gain and loss" 617 model. The approach must be applied to subsections of relatively small size (<1 or 2 m 618 of thickness), so that the model assumptions can be met, especially those concerning 619 the constancy of the mobility parameters along the weathering zone. The results also 620 confirm that the upper part of the weathering profile, influenced by 621 surface/vegetation/biological processes, is not well adapted for applying the U-series 622 nuclides methodology for recovering regolith production rates. Furthermore, they 623 624 indicate that in the deeper parts of the weathering profile, all parts of the profile are not necessarily appropriate for the application of the modeling approach, especially in the 625

262 zones marked by short distance variations of mineral paragenesis.

As illustrated with the current study of the Longnan weathering profile, 627 independent determinations of the regolith production rates on different subzones of the 628 weathering profile proves to be a relevant approach to assess the long-term stability of 629 the regolith production. Thus, for the Longnan profile, the observation of similar values 630 of ~ 2 m/Ma in each of the four sub-zones where the regolith production rate has been 631 independently determined, suggests that such a profile of more than 5 million years has 632 633 been formed at a relatively stable long-term production rate. Such a stable and slow regolith production rate of 2 m/Ma at depth can be reconciled with the previously 634 published in situ ¹⁰Be data from the upper part of the same profile, when assuming no 635 steady-state erosion of the upper part of the profile. In this case, the ¹⁰Be concentrations 636 near the top of the profile result from both a recent stabilization during 60 ky at least 637 and an inherited component of $20*10^3$ atoms/g accumulated at great depth. 638 Accumulation of ¹⁰Be at depth is due to muonic production during long stable periods 639 of time and brought near the surface due to exhumation of the deeper part of the profile 640 641 as a result of short periodic intense erosional events.

Altogether our study demonstrates that the combined analysis of U-series and cosmogenic nuclides, which has the potential to become a relevant approach to constrain the dynamics of continental surfaces, requires (a) dense and deep sampling for both nuclides studies, and (b) also to consider more systematically the polyphased history of erosion of the continental surface during the Quaternary, i.e. the long-term accumulation of ¹⁰Be at depth due to muonic production.

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- 660 **Table captions:**
- 661 Table 1: U, Th concentrations and $(^{234}U/^{238}U)$, $(^{230}Th/^{234}U)$, $(^{230}Th/^{232}Th)$ activity ratios 662

Table 2: Production rates and mobility parameters (ki, fi) obtained for zones 2, 3a, 4a,
and 4b (see text for details).

665

666 **Figure captions**:

Figure 1: A) Location of study site in southeast China on mean annual precipitation
map of Asia for period 1961-1990 (Schamm et al., 2014; http://gpcc.dwd.de). Arrows
are main moisture sources. B) Location of weathering profile (red dot) near Taojiang
river left bank north of Longnan city (image background from Google Earth). C) Field
view of soil profile outcrop at bottom of forested ridge, people for scale. Inset shows
numbered positions of samples. Sample thickness is 10, 20 and 50 cm between 0-1, 18, and below 8 m, respectively.

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Figure 2: Depth variation of K-feldspar and plagioclase abundances along weathering profile, together with chemical index of alteration (CIA), K/Ti, and grain size (data from Zhang et al. (2015) and Liu et al. (2016)). Samples analyzed in this study and in different zones are shown on schematic column to right, with quartz and plagioclase veins depth indicated. Grey boxes are sample position, black squares are previously analyzed and published samples (Zhang et al., 2015; Liu et al., 2016).

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Figure 3: Variations of U (ppm), Th (ppm), U/Th (ppm/ppm) and Th/Ti (ppm/ppm)
with depth with location of modeling zones. Error bars are 2%, 1.3%, <1.6%, < 5.5%
from left to right. U-Th Data in Table 1, Ti data from Zhang et al. (2015).

- Figure 4: Depth variations of $(^{234}U/^{238}U)$, $(^{230}Th/^{234}U)$ and $(^{230}Th/^{232}Th)$ ratios with location of modeling zones. For uncertainties see Table 1.
- 688

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Figure 5: (²³⁰Th/²³⁴U) vs (²³⁴U/²³⁸U) diagram for zones 2, 3a, 4a, and 4b. Grey curve is
best-fit calculated with model described in text for each zone. Sample ages are relative
to deepest sample in each zone.

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Figure 6: gain (k_i) and loss (f_i) coefficient ratios, k_{234}/f_{234} as a function of k_{238}/f_{238} for model results of zones 2, 3a, 4a and 4b. Inset is zoom for ratios below 1

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Figure 7: A) In situ ¹⁰Be concentration in guartz (red cross; data in Cui et al., 2016) and 696 modeled exponential decrease with depth. green and blue lines are modeling following 697 Yang et al. (2019), blue assumes steady-state and no inheritance, green with inheritance. 698 Local ¹⁰Be spallogenic production rate of 3.74 atoms/g/yr from CRONUS online 699 calculator (Marrero et al., 2016). Slow and fast muons production rate of 0.1066 and 700 0.0485 atoms/g/yr, respectively (Yang et al., 2019). B) Grey boxes (below 2 m depth) 701 are U-series regolith production rates, green and blue boxes (from surface to 4 m depth) 702 703 are ¹⁰Be denudation rates with inheritance and without inheritance, respectively (see

text for details).

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Figure 8: Long term evolution of a weathering profile modulated by high and low frequency climate changes as depicted with benthic oxygen isotope curve over 800 ka (top) and 5.5 Ma (bottom), respectively (Lisiecki and Raymo, 2005). E₁ and E₂ are longand short-term surface denudation rates. P is steady-state production rate of regolith at depth. Profile evolution steps as follows: 1: Steady state; 2: Fast erosion over a short period; 3: Return to steady state; 4: Steady state.

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713 References:

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) 2800 Time (ka) climatic modulation

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Table 1: U, Th concentrations and (²³⁴U/²³⁸U), (²³⁰Th/²³⁴U), (²³⁰Th/²³²Th) activity ratios

JLN-S1 Sample Number	Depth (cm)	TiO ₂ (wt%)	$\begin{matrix} [U]\\(mg/kg)\\\pm 2\%\\(2\sigma)\end{matrix}$	[Th] (mg/kg) ±1.3% (2\sigma)	(²³⁴ U/ ²³⁸ U)	±2σ	(²³⁰ Th/ ²³⁴ U)	±2σ	(²³⁰ Th/ ²³² Th)	±2σ
03	30-40	0.43	5.89	53.8	1.009	0.004	1.032	0.011	0.349	0.003
05	50-60	0.31	4.40	39.0	1.011	0.004	1.034	0.011	0.361	0.004
07	70-80	0.24	3.55	30.5	1.005	0.004	1.036	0.011	0.371	0.004
09	90-100	0.22	3.31	27.4	1.012	0.004	1.038	0.011	0.388	0.004
11	110-120	0.24	3.77	30.5	1.014	0.004	1.048	0.012	0.401	0.004
13	130-140	0.23	4.12	31.5	0.994	0.011	0.998	0.011	0.397	0.004
15	150-160	0.31	5.04	40.6	1.003	0.004	0.966	0.011	0.368	0.004
20	200-220	0.25	4.17	32.9	0.990	0.004	0.995	0.017	0.382	0.005
23	260-280	0.24	4.22	31.4	1.008	0.004	0.945	0.010	0.392	0.004
25	300-320	0.27	5.29	34.7	1.008	0.011	0.881	0.010	0.414	0.004
26	320-340	0.26	5.55	37.3	1.014	0.004	0.890	0.010	0.411	0.004
27	340-360	0.23*	4.93	37.1	0.997	0.011	0.908	0.010	0.368	0.004
28	360-380	0.29	8.55	40.8	0.990	0.011	0.853	0.009	0.541	0.005
29	380-400	0.26	6.63	32.3	0.978	0.004	0.814	0.009	0.500	0.005
33	460-480	0.28	5.98	43.0	0.978	0.004	0.926	0.016	0.385	0.005
36	520-560	0.21	4.99	28.1	1.000	0.004	0.970	0.011	0.527	0.005
37	560-580	0.26	6.12	32.5	1.005	0.004	0.932	0.016	0.540	0.007
39	600-620	0.23*	3.63	31.7	0.988	0.011	1.012	0.011	0.350	0.004
41	640-660	0.21	4.58	27.4	0.989	0.004	0.910	0.010	0.460	0.005
42	660-680	0.28*	5.74	44.2	0.976	0.011	0.974	0.026	0.378	0.009
43	680-700	0.21*	3.98	31.8	0.965	0.011	1.042	0.011	0.385	0.004
44	700-720	0.23	3.82	33.6	0.976	0.004	1.081	0.012	0.367	0.004
46	740-760	0.23*	4.39	36.9	0.980	0.011	1.074	0.012	0.383	0.004
47	760-780	0.26	3.75	31.8	0.957	0.004	1.122	0.019	0.388	0.005
49	800-850	0.17*	3.50	30.3	0.936	0.010	1.043	0.011	0.346	0.003
50	850-900	0.24	3.61	34.4	0.937	0.004	1.095	0.012	0.330	0.003
51	900-950	0.23	3.82	35.3	0.948	0.004	1.095	0.019	0.344	0.004
52	950-1000	0.21	3.77	30.4	0.967	0.004	1.069	0.012	0.392	0.004
53	1000-1100	0.18	3.19	29.8	0.941	0.004	1.090	0.019	0.335	0.004

Uranium concentrations and activity ratios were calculated assuming an isotopic 238 U/ 235 U ratio of 137.83 (Weyer et al., 2008) and decay constants λ_{238} and λ_{234} of 1.551×10^{-10} yr⁻¹ and 2.826×10^{-6} yr⁻¹, respectively (Jaffey et al., 1971; Cheng et al., 2000), and λ_{230} and λ_{232} of 9.158×10^{-6} yr⁻¹ and of 4.932×10^{-11} yr⁻¹, respectively (Cheng et al., 2000; Rubin, 2001). Analytical uncertainties ($\pm 2\sigma$) were estimated from the 2SD-reproducibility observed for all the sample powder duplicates (n=7) and the rock standard replicates (n=4). For some measurements this global estimation was adjusted to account for the internal error of the measurement or to the result of the rock standard of the session. TiO₂ (weight percent) concentrations are from Liu et al. (2016) and Zhang et al. (2015) except (*), which has been analyzed for this study at the China University of Geosciences (Wuhan) following the approach given in Zhang et al. (2015). Precision on Ti concentrations are estimated to be $\pm 2\%$.

Table 2: Production rates and mobility parameters (ki, fi) obtained for zones 2, 3a, 4a, and 4b (see text for details).

Zone	Var.	P (m/Ma)	$k_{238}(y^{-1})$	$f_{238}(y^{\text{-}1})$	$k_{234}(y^{-1})$	$f_{234}(y^{-1})$	k_{234}/k_{238}	f_{234}/f_{238}	k_{238}/f_{238}	$k_{234}\!/f_{234}$
Zone 2	mean	2.21	5.37 E-07	17.4 E-07	9.60 E-07	24.9 E-07	1.98	1.44	0.32	0.42
	Std dev.	0.21	1.94 E-07	2.3 E-07	2.94 E-07	3.15 E-07	0.40	0.14	0.11	0.11
Zone 3a	mean	2.66	8.79 E-07	40.6 E-07	9.14 E-07	23.6 E-07	1.35	1.15	0.33	0.38
	Std dev.	0.23	1.70 E-07	2.24 E-07	2.34 E-07	3.10 E-07	0.26	0.11	0.07	0.07
Zone 4a	mean	1.85	24.2 E-07	9.92 E-07	30.5 E-07	14.3 E-07	1.29	1.51	2.54	2.22
	Std dev.	0.59	11.4 E-07	7.81 E-07	16.3 E-07	11.3 E-07	0.14	0.26	0.84	0.68
Zone 4b	Mean	2.33	12.4 E-07	5.23 E-07	17.9 E-07	7.21 E-07	1.52	1.61	2.38	2.44
	Std dev.	0.23	2.73 E-07	2.00 E-07	3.11 E-07	2.42 E-07	0.30	0.56	0.47	0.50