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1	Comment on "First records of syn-diagenetic non-tectonic
2	folding in Quaternary thermogene travertines caused by
3	hydrothermal incremental veining" by Billi et al.
4	Tectonophysics 700-701 (2017) 60-79
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#### 38 Abstract

39 Billi et al. (2017) proposed a new interpretation for the origin and internal structure of thermogene travertine deposits. On the basis of evidence from two 40 quarries located in southern Tuscany (Italy), they interpreted some travertine 41 42 beds as calcite veins and argued that undulating travertine beds formed by syn-diagenetic (i.e. non-tectonic) folding that was caused by laterally-confined 43 volume expansion caused by incremental veining. They assumed that such a 44 45 process causes changes to the rock properties, including porosity reduction, rock strengthening, and age rejuvenation. The interpretations by Billi et al. 46 (2017) challenge and question the current understanding and interpretation of 47 thermogene travertine deposits. This understanding, based on numerous 48 studies since the 1980s, is that these deposits form from thermal water flowing 49 downslope, and precipitating calcium carbonate. Here, we explain how the 50 51 comparison with active depositional systems is essential for the understanding the origin of structures in older, inactive travertine deposits, such as those 52 studied by Billi et al. (2017). We further argue that the three-dimensional 53 54 setting of travertine deposits should be taken into account in order to discuss 55 the possible development of secondary structures. Indeed travertine deposition on slopes typically leads to the formation of terraced morphologies with pools 56 57 bordered by rounded rims and separated from each other by steep walls. The resulting three-dimensional structures can be misinterpreted as asymmetric 58 folds in two-dimensional views (i.e., in saw-cut walls of quarry). In this paper 59 we debate the interpretations offered by Billi et al. (2017) and their criteria to 60 recognise syn-diagenetic, non-tectonic folds in travertine deposits, and explain 61 why many of their ideas are questionable. 62

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64

#### 65 Key words

travertine facies, travertine depositional geometry, deformational processes, calcite veins,
 enterolithic structures, age rejuvenation

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## 70 **1. Introduction**

The recent literature on travertine (i.e. thermogene terrestrial carbonate) has 71 made use of this deposit as a proxy for palaeo-environmental (Bertini et al. 72 2008; Ricci et al., 2015) and climate change reconstructions (Sturchio et al., 73 1994; D'Argenio et al. 1995; Rihs et al., 2000; Soligo et al., 2002; Mesci et al., 74 2008; Faccenna et al., 2008; Zentmyer et al., 2008; Sierralta et al., 2010; 75 Brogi et al., 2010), neotectonic and palaeoseismological analyses (Altunel and 76 Hancock, 1993a, 1993b; Çakır, 1999; Hancock et al., 1999; Brogi, 2004; 77 Altunel and Karabacak, 2005; Uysal et al., 2007; 2009; Mesci et al., 2008; 78 Brogi and Capezzuoli, 2009, 2014; Temiz et al., 2009; 2013; Brogi et al., 79 2010, 2012, 2014a, 2014b, 2016; Altunel and Karabacak, 2005; Uysal et al., 80 2007; 2009; Hancock et al., 1999; Temiz et al., 2009; 2013; Temiz and 81 Eikenberg, 2011; Cakir, 1999; Mesci et al., 2008), geothermal exploration 82 (Navarro et al., 2011; Pasvanoğlu and Chandrasekharam, 2011; Alçiçek et al. 83 2016; Brogi et al., 2016; Alçiçek et al., 2017), elemental biomediation 84 processes analyses (Folk, 1994; Bonny and Jones, 2003; Fouke et al., 2003; 85 Rogerson et al., 2014), and natural CO<sub>2</sub> degassing evaluation (Shipton et al., 86 2005; Uysal et al., 2011; Frery et al., 2016). These applications coupled with 87 88 the fact that travertine is a rare carbonate deposit, makes it a precious archive of information from many different scientific perspectives. The conventional 89 approach for the study of these deposits requires many different mandatory 90 steps, including: (i) reconstruction of the three-dimensional geometry of the 91 travertine deposit and its evolution through time; (ii) reconstruction of the 92 depositional architectural setting of the different depositional stages; and (iii) 93 94 sedimentary facies analysis that includes interpretation of sedimentary facies, their lateral relationships, and the processes associated with each depositional 95 setting. The latter step is critical because it allows the reconstruction of the 96 environmental features that controlled the travertine formation and the related 97 sedimentary processes dictating its origin. Accurate facies interpretation 98 depends on careful observations that follow a well-established methodology, 99 which has been fully documented in a considerable number of previous studies 100 (e.g. Chafetz and Folk, 1984; Guo and Riding, 1992, 1994, 1998; Chafetz and 101 Guidry, 1999; Pentecost, 2005; Jones and Renaut, 2010; Gandin and 102

103 Capezzuoli, 2014).

Failure to follow the established protocols can lead to misinterpretations. This 104 is, in our opinion, the case presented in the paper by Billi et al. (2017) who 105 106 analysed Pleistocene (thermogene) travertine deposits exposed in saw-cut 107 walls of two quarries (see also: Ronchi and Cruciani, 2015), located in southern Tuscany (Italy). They interpreted structures in those walls as syn-108 109 diagenetic, non-tectonic veins and folds and provided nine criteria for discriminating secondary structures (i.e. post-depositional syn-diagenetic 110 processes such as veining, folding, or rejuvenation actions) from primary 111 112 structures (i.e. related to the sedimentary evolution) in travertine deposits. We question these criteria, which in our view do not sufficiently take in to account 113 the present knowledge on travertine formation. Herein, our comments aim to 114 115 fill this gap and to favour a solid consideration of the comparison between 116 active and fossil travertine depositional systems.

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### 118 **1.1 Questions for comments**

119 Billi et al. (2017, their Figs. 3, 4) argued that the syn-diagenetic (i.e. non-120 tectonic) folding of travertine was the result of laterally-confining volume expansion that was caused by incremental hydrothermal veining. Their idea, 121 122 based on the study of Gratier et al. (2012), implied that undulating travertine beds are unreliable indicators of the sedimentary environment (and its 123 evolution through space and time) in which the travertine accumulated. 124 125 Furthermore, a consequence of their hydrothermal-veining interpretation is age rejuvenation of travertine deposits with implications for geochronological 126 results and modification of travertine strength and porosity, and effective 127 impacts on permeability evaluations. On the basis of these considerations, Billi 128 et al. (2017) proposed "a list of significant criteria to discriminate secondary 129 from primary structures and to identify rejuvenation processes" to explain the 130 131 occurrence of: (i) radiometrically-dated structures that are younger than 132 overlying ones; (ii) downward growth of crystals; (iii) veins overprinting/cutting through overlying or underlying beds; (iv) relicts of 133

134 primary porous travertine beds between radiating vein crystals; (v) increasingly deformed primary structures such as beds or pores towards veins 135 and folds; (vi) bed-normal foliations and second-order folds nested inside 136 larger folds; (vii) polyphase folding including overturned folds with refolded 137 limbs; (viii) stylolite surfaces parallel and stylolite teeth normal to vein planes; 138 (ix) post-depositional non-karstic voids between folded and flat veins and 139 140 beds, outlining the occurrence of post-depositional detachment mechanisms between adjacent beds. We criticize the interpretation of these points. The 141 following text is therefore organized in separate sections, each of which 142 focuses on those aspects of the interpretations that are crucial for a well-143 constrained facies analysis and reconstruction of the 144 depositional environments. In so doing, we underline and stress the lessons that have been 145 146 learnt from the analysis of active travertine depositional systems, which 147 collectively provide clear insights into the processes that define the depositional geometry and development of travertine deposits. 148

149

### 150 **2.** Calcite veins versus travertine beds/layers (crystalline crusts)

151 This section discusses the points 2 to 5 of the list of criteria provided by Billi et 152 al. (2017) for the interpretation of travertine deposits.

Billi et al. (2017) described the geometric relation between the porous 153 travertine beds and the growth direction of the constituent calcite crystals (Billi 154 155 et al., 2017, their Figs. 4d, 4e and 11d) in the crystalline travertine beds (also termed crystalline crusts by Guo and Riding, 1998 and references therein). 156 Based on this, Billi et al. (2017) argued that the porous travertine beds formed 157 as a primary deposit, whereas the crystalline crusts developed as calcite veins 158 159 that post-dated deposition of the travertine. This interpretation challenges the fact that crystalline crusts are primary carbonate precipitates that develop at 160 161 the depositional surface as a result of  $CO_2$  degassing from carbonate-rich waters during their flow (Fig. 1). Thus, crystalline crusts are formed as a 162 variety of calcite/aragonite crystals (feather, fan, dendrites; see Jones and 163 Renaut, 1995; 2010 for a review; Jones et al., 2000, 2005). Furthermore, this 164 clarifies the definition of travertine as a primary bedded thermogene deposit 165

166 (Chafetz and Folk, 1984; Jones and Renaut, 1995, 2008; Jones et al., 1996, 2000, 2005; Guo and Riding, 1998; Jones and Renaut, 1995, 2008; Jones et 167 al., 1996, 2000, 2005; Rainy and Jones, 2009; Gandin and Capezzuoli 2014; 168 Della Porta, 2005; Croci et al., 2016; Della Porta et al., 2017) where the 169 170 intercalation of porous (Fig. 2) and crystalline deposits is a characteristics of 171 travertine spring deposits (Riding, 1991; Pedley, 1990; Flügel, 2004; 172 Pentecost, 2005; Pedley, 2009; Brogi et al., 2010; Capezzuoli et al., 2014; Pola et al 2013; Gandin and Capezzuoli, 2014; Gradzinski et al., 2014). 173

Veins (see Bons et al., 2012 for a review) in travertine deposits (**Fig. 3**) are formed of different types of crystals (Altunel and Karabacak, 2005; Uysal et al., 2009, 2011; Rimondi et al., 2015; Brogi et al., 2014a; Brogi et al., 2016) and fill cracks that cut across layers (**Fig. 3a-f**) or follow bedding surfaces (**Fig. 3g-i**) (e.g. Altunel and Hancock, 1993a, 1993b; Altunel and Karabacak, 2005; Mesci et al., 2008; Uysal et al., 2009; 2011; Brogi et al., 2016; Brogi et al., 2017; Selçuk et al., 2017).

181 The growth of bed-parallel veins (i.e. sub-horizontal veins that opened against the force of gravity) has been attributed to: i) the crystallization force of calcite 182 triggered by  $CO_2$  degassing at depths of 1–10 m (Gratier et al., 2012); ii) 183 184 repeated injections of high-pressure hydrothermal fluids (pressure exceeding the weight of the overlying rocks volume, Brogi et al., 2016) during seismic 185 events (Uysal et al., 2007; Altunel and Karabacak, 2005; Uysal et al., 2007; 186 Brogi and Capezzuoli, 2014; Brogi et al., 2017) and/or (iii) by climate induced 187 pressure variations within the geothermal reservoir at depth (Uysal et al., 188 189 2009). The textures of such veins (Fig. 4), however, are completely different from the ones reported by Billi et al. (2017, their Fig. 4e). In their case, the 190 veins are formed of needle-like crystals, rows of palisade, fibrous or prismatic 191 crystals, generally arranged in tight palisades with straight extinction or 192 193 clusters of ray-shaped fans (as described in Folk et al., 1985; Atabey 2002; see also Flügel, 2004, Jones and Renaut, 2010, Gandin and Capezzuoli, 2014, 194 for review on the petrographic characteristics of calcite veins and travertine). 195 196 They are typical of precipitates associated with pools and terraces (Figs 1 and 197 2). Such crystalline layers typically consist of dense crystalline dendrites or 198 crusts (some could also be shrubs as illustrated in Tivoli, nearby Rome, Italy,

as documented by Erthal et al., 2017) that can laterally, or vertically, grade 199 into the porous deposits where numerous voids from bubbles and other origins 200 are present (Fig. 2). This arrangement is readily evident in present-day active 201 travertine deposits all over the world and defines well-documented depositional 202 geometries such as dams, rims, and terraces (e.g. Pamukkale, Turkey; 203 Mammoth Hot Springs, USA; Huanglong, China; Badab-e surt, Iran, Saturnia, 204 205 Italy; Rapolano Terme, Italy; see also Pentecost, 1995, 2005; Ford and Pedley, 1996 for a review). Similar depositional structures are present in caves, where 206 the same process can be active (rimstone dams; Ford and Williams 2007). 207

These well-documented relationships between porous and crystalline beds 208 (Figs 2 and 5 for active and fossil examples, respectively) also include what 209 Billi et al. (2017, their Fig. 5a and 5b) described as "chimney-like veins". In 210 211 their photographs, it is apparent that the brownish porous layers pass laterally into the dense crystalline layers ("veins" in Billi et al., 2017), with visible 212 interfingering relationships. This cannot, however, be attributed to a 213 subsequent deformational event. Moreover, their Fig. 4f shows that the 214 boundary of the crystalline beds with the porous micritic deposits is diffuse, 215 underlining the absence of mechanical/physical discontinuities that would be 216 217 expected if this were caused by subsequent pressure-induced deformation as proposed by Billi et al. (2017). 218

Finally, Billi et al. (2017) based most of their arguments on evaluation of the direction of calcite crystals growth. Conversely, the criteria used to establish the growth-orientation of the travertine beds should be considered with care, because the fan-like crystal arrangement (**Fig. 1d-g**) may result in different apparent directions within the same bed (e.g. Billi et al. 2017, their Figs S25 and S34).

Billi et al. (2017) suggested that the development of calcite veins caused the reduction of primary porosity (their Fig. 5c), as indicated by the flattened pores, which they assumed were originally almost spherical (see their section 3.1.3). It is important to stress, however, that the classical, spherical "coated bubbles" found in many travertine deposits (Guo and Riding, 1998; Gandin, 2013; Gandin and Capezzuoli, 2014 for a review) are formed by the encrustation of bubbles with a thin coating of calcite caused by CO<sub>2</sub> degassing

(see Schreiber et al., 1981; Chafetz et al., 1991 and Pentecost, 2005 for a 232 complete description of this depositional process). When this is the case, the 233 bubbles are externally characterized by thin calcite rims. Consequently, this 234 type of porosity is commonly concentrated in small volumes (Fig. 2). 235 Otherwise, most pores in travertine (as all the pores discussed in Billi et al., 236 2017) are formed from photosynthetic oxygen that is produced by microbial 237 activity and are common in microbial deposits (e.g. Folk et al., 1985; Riding, 238 1991; Rainey and Jones, 2009). Given that these are not coated by a calcite 239 rim, their shape is highly variable (e.g. Gandin and Capezzuoli, 2014 for a 240 comparison) and reflects the balance among gas pressure, gravity, and weight 241 of the microbial mat. Thus, the pore shape cannot be used to evaluate 242 deformation in travertine deposits, because their initial shape is highly variable 243 244 and unpredictable. It follows that the interpretation of the crystalline beds as calcite veins that postdated the original travertine deposits is difficult to 245 246 accept.

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## **3. 2D-view of crystalline crusts** *versus* **folded travertine**

Hereafter we discuss the points 6 and 8 of the list of criteria that Billi et al.(2017) proposed for the interpretation of travertine deposits.

Billi et al. (2017) argued that undulations in the travertine beds are syn-252 diagenetic folds that developed during the progressive and incremental 253 formation of syntaxial, bedding-parallel, calcite veins. In active spring 254 depositional environments (Fig. 1) the undulation in travertine deposits 255 256 (undulated layers) is a natural consequence of subaerial deposition on variably inclined surfaces (i.e. slopes and dammed zones), through pools and rims, 257 producing terraced surfaces (Fig. 2). This is well constrained by three-258 dimensional observations in most active and fossil travertine (and tufa) 259 depositional systems (Italy: Chafetz and Folk, 1984; Guo and Riding, 1998; 260 D'Argenio et al., 1981, D'Argenio and Ferreri, 1987,1988; Hungary: Scheuer 261 262 and Schweitzer, 1981; Kele et al., 2008; Claes et al., 2017, Török et al., 2017; Turkey: Altunel and Hancock, 1993a; 1993b; Khatib et al. 2014; Lebatard et 263 al., 2014; Claes et al., 2015; Tunisia: Henchiri et al., 2017; China: Liu et al., 264

1995; Lu et al., 2000; Central Italy: Capezzuoli et al., 2014; Della Porta,
2015; Croci et al., 2016; Della Porta et al., 2017; Violante et al., 1994a,
1994b; Central Western Carpathians, Slovakia: Gradzinski et al., 2014; USA:
Fouke et al., 2000; different places: Pentecost 2005; Alonso-Zarza and Tanner
2010; Arenas-Abad et al. 2010; Jones and Renaut 2010).

Taking a different viewpoint, Billi et al. (2017), evaluating geometries based on 270 2D-cross sections, interpreted the undulatory and terraced travertine beds as 271 asymmetric folds, and argued that the typical aggradational and slightly 272 progradational geometry of the pool/rims are the result of second-order folds. 273 They did not consider the well-established fact that travertines are self-274 regulating systems, that can modify their own depositional environment, which 275 276 may result in changes in the attitude of the strata with, or even without, any 277 syn- or post-depositional tectonic deformation.

Ronchi and Cruciani (2015), who studied the same quarries as Billi et al. 278 (2017), interpreted the terraced travertine as an effect of the travertine 279 deposition on pre-existing slopes. Billi et al. (2017) did not provide convincing 280 reasons for modifying the interpretations proposed by Ronchi and Cruciani 281 (2015) and the numerous of studies from other areas that show the same 282 features (Chafetz and Folk, 1984; Guo and Riding, 1998: D'Argenio et al. 283 1981; Scheuer and Schweitzer, 1981; Altunel and Hancock, 1993a; Khatib et 284 al. 2014; Lebatard et al., 2014; Claes et al., 2015; Henchiri et al., 2017; Liu et 285 al., 1995; Lu et al., 2000; Pentecost, 2005 for a review; Capezzuoli et al., 286 2014, Della Porta, 2015; Croci et al., 2016; Della Porta et al., 2017). 287

Billi et al. (2017, their Figs 5f and 5g) also described folds with evident hinge 288 289 thickening that affect the calcite veins. They proposed for a composite deformational process, with initial formation of mostly sub-horizontal calcite 290 veins that was followed by folding of the previously developed veins and 291 parallel porous travertine beds. They also argued that there was a sub-292 horizontal tectonic foliation developed in association with the second-order 293 folds developed only in the vertical fold limb of the first order structures (see 294 295 their Fig. 5g and 6). In addition, their Fig. S10 is meant to show refolded structures resulting from at least two folding events, with deformation of the 296 limb of a larger overturned fold. 297

The folds, refolded folds, and related tectonic foliation proposed by Billi et al. 298 299 (2017) have already been unequivocally described as primary depositional features (cf. Chafetz and Folk, 1984; Hammer et al., 2010), but they did not 300 discuss or refute this interpretation. The undulatory beds that they interpreted 301 as first-order folds (and refolded folds) are explained as an effect of the plane 302 view orientation in a travertine cascade environment (Fig. 6a-b). Their 303 "second order folds" and associated "axial planar tectonic foliations" (cf. their 304 Figs 6b, 6d and 6f) can be explained as microterraces (Fig. 6c-e) that formed 305 on spring slopes (e.g. "rimstone pool" of Warwick, 1952, to "terracette" of 306 Bargar, 1978 and Guerts et al., 1992; "minidams" of Pentecost 2005 and Jones 307 and Renaut 2010; "microterracettes" in Hammer et al., 2010). 308

The 2D-features displayed on the quarry walls by Billi et al. (2017, their Fig. 5 309 310 and 6) are recognizable in all present-day depositional systems (Fig. 2). This circumstance reinforces the argument that these are really vertical cross-311 sections through fossil pool rims (i.e. microterraces) and cascades that formed 312 by natural (i.e. primary) sedimentary processes (Fig. 6); they are not 313 (second-order) folds as suggested by Billi et al. (2017). In addition, it should 314 also be remembered that the shape, size, and distance between micro- and 315 macro-terraces depend on local depositional features (e.g. Riding, 1991; 316 Pentecost 2005: see Fig. 16; Goldenfeld et al., 2006; Hammer et al. 2005, 317 2007, 2010; Veysey and Goldenfeld, 2008), not only for travertine, but also for 318 all flowstone deposits (speleothems, tufa, siliceous sinter). 319

Billi et al. (2017) claimed a further explanation for their minor folds. They 320 suggested that the process that produced the small-scale folding is analogous 321 322 to the process that produces enterolithic and/or tepee structures, which are commonly found in evaporitic deposits. Enterolithic structures (i.e. irregular, 323 highly-non-cylindrical tight to open folds) are typical of sabkha environments 324 and cannot develop in travertine deposits. Such structures are produced by 325 localized changes in volume after evaporite deposition (cf. Gandin and Wright, 326 2007). This syn- to meta-depositional deformation is induced by the chemical 327 328 transformation of the sulphates, such as the swelling of anhydrite during hydration to gypsum. Gypsum and anhydrite nodules form through the 329 330 capillary system within the upper phreatic zones beneath the sabkha surface,

331 displacing and replacing sediment under pressurized saline fluids that are flowing through pores due to evaporative capillarity (Tucker, 1988; Warren, 332 1999, 2006; Flügel, 2004; Gandin and Wright, 2007). These intrasediment 333 334 crystals grow in a matrix of fine sediment (i.e. lime mud, clays), where the nodules grow and coalesce to form the enterolithic structures. Thermogene 335 336 travertine, however, with primary precipitation of calcite/aragonite, has a 337 completely different chemical and mineralogical composition, as well as an internal organisation, and its deposition occurs in totally different 338 339 environments. Thus, there is no basis for assuming that travertine deposits may develop in the same manner as evaporites and therefore, extreme care 340 should be taken when comparing the two. 341

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#### 343 **3.1 Mechanics of travertine folding**

This section discusses the points 7 and 9 of the list of criteria proposed by Billi et al. (2017).

Travertine forms at the subaerial surface, where deposition and lithification 346 processes are almost contemporaneous (Pentecost, 1995). Travertine is 347 composed largely of calcite and/or aragonite, carbonate minerals that are 348 brittle under low temperature conditions (< 250°C, Rutter, 1972). 349 350 Furthermore, the maximum thickness of travertine in slope environments does 351 not exceed a few tens of meters as found in several areas worldwide (Pentecost 1995; Guo et al., 1996; Hancock et al., 1999; Brogi, 2004; Brogi et 352 al., 2010; Ronchi and Cruciani, 2015; Khatib et al. 2014; Lebatard et al., 353 2014; Claes et al., 2017). This implies that travertine can only be affected by 354 355 deformation at, or near the surface. It follows that the boundary conditions (P, 356 T and/or significant content of interstitial fluids) required to obtain highly non-357 cylindrical folds (Billi et al., 2017, their Fig. 5j) are not present. In this view, the flattened, squeezed, dragged pores as Billi et al. (2017) described close to 358 the fold hinges, veins and folded veins (Figs. 3b, 5c, d, S6, S11, S12, S14, 359 S23, and S35), are not the result of the folding mechanism but are primary 360 sedimentary features, as discussed above. 361

362 Second-order folds attributed to folding imply bedding viscosity contrast 363 and/or thickness variations of a flexural slip folding mechanism. This is

normally produced in a multi-layered succession, which does not seem to apply 364 to travertine formed by carbonate precipitation on a terraced slope. In order to 365 explain fold hinge thickening and occurrence of second-order folds and 366 foliations, Billi et al. (2017, their Figs. 5f, g, and 6) considered the Biot-367 Ramberg's buckling equation (their Eq. 1). Billi et al. (2017) used the buckling 368 equation for single-layer folding and an assumed Newtonian viscosity to obtain 369 a viscosity contrast of about 1.5 to 4. They did not report the amplitudes of the 370 folds (A) in relation to the fold wavelength ( $\lambda$ ), which is shorter than the arc 371 length (L) that they used. Although it is difficult to ascertain how layer 372 thicknesses (t) were defined in the multi-layer setting, it seems that a ratio of 373 374  $A/I \ge 0.25$  is a conservative estimate for the structures. Their reported L/t ratios range from about 4 to 5. This can be converted to t/l ratios, using  $l \ge 2/3 \cdot L$ , in 375 the range of 0.1 to 0.25. 376

 $A/\lambda$  versus  $t/\lambda$  trends depend on the amount of shortening and viscosity 377 contrast (Fig. 6 in Schmalholz and Podladchikov, 2001; Fig. 2 in Llorens et al., 378 2013). For single-layer folding, the range of measured ratios would indicate 379 380  $\geq$ 50% shortening and a viscosity ratio between 25 and 250. At the very low viscosity contrast, as used by Billi et al. (2017), folds grow very slowly in 381 amplitude as layer thickening dominates. To achieve  $A/\lambda >>0.25$ , a very high 382 383 strain would be required, but this would result in much higher  $t/\lambda$  ratios (>>0.5) than the ones observed. 384

If the buckle fold theory were applicable (which we doubt), the viscosity 385 contrast between the layers in the travertine would need to be much higher 386 than that proposed by Billi et al. (2017). More importantly, the amount of 387 strain needed to achieve the high  $A/\lambda$  ratios would be well over 50% 388 389 shortening, or in case of constrained volume increase, >100% of layer length increase by volume change. In addition, Billi et al. (2017, their Fig. S11) also 390 described bedding-parallel stylolites that are typically associated with 391 dissolution that takes place as a result of overburden loads during deep burial 392 and pressure solution (e.g. Bathurst, 1995; Rolland et al., 2012; Heap et al., 393 2014; Koehn et al., 2016). Their development is related to the occurrence of 394 local rock mass heterogeneity, occurrence of interlayered/intracrystalline 395 396 water, dissolution and deposition of dissolved material in extensional veins,

and a sufficient lithostatic pressure. As shown by Sellier (1979), the pressure-397 dissolution of calcite begins at a depth of about 300 m. In other cases, 398 stylolites have been documented at lesser burial depth, at about 90 m, but 399 only if the limestones have a high clay content (Schlanger, 1964). Taking into 400 account that the travertine deposits described by Billi et al. (2017) were not 401 402 buried to the depth needed for stylolite formation, we doubt that the structures described by Billi et al. (2017) are stylolites. The 2D-view of the outcrop is 403 again at the base of this misunderstanding: features similar to stylolites can be 404 produced if the saw-cut walls are about orthogonal (or at high angle) to the 405 406 crystalline crust on slopes that were originally terraced (**Fig. 6c-e**).

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### 408 **4. Travertine age rejuvenation**

This part is dedicated to the discussion of the point 1 of the list of criteria provided by Billi et al. (2017), concerning the interpretation of the age of travertine deposits.

Radiometric age dating of travertine is possible by analysis of the U/Th content 412 in the calcite molecules (Taylor and McLennan, 1995 and references therein). 413 U/Th dating of carbonates, younger than 500ka (Walker, 2005), is a sensitive 414 analysis that relies on carefully collected samples (Ku and Liang, 1985). The 415 best dates come from compact, non-porous samples (cf. Carrara et al., 1998; 416 Brogi et al., 2010). Porous travertine samples can be problematical because 417 younger calcite and/or aragonite cements found in the pores may have formed 418 419 at any time after deposition. The presence of these cement phases can seriously affect the isotopic and geochronological results. Billi et al. (2017, Fig. 420 7) showed inconsistent age-dating results through a travertine section 421 encompassing "veins" and "porous" travertine. The "inconsistent" ages 422 (samples CP15\_8, CP14\_2, ST1 and ST3) were used as criteria to validate the 423 "vein" formation. Two samples of travertine layers (lower photo in their Fig. 424 425 7b) show an inverted age order, whereas, other samples (upper photos in their Fig. 7b) show upward younging. Following the principle they adopted, 426 however, only one "vein" sample (CP15\_8, at the lowest position) is younger 427

than the sample that represents the supposed hosting travertine (CP14 25), 428 whereas the other "veins" produced older (CP15 1; CP14 5), or coeval 429 (considering the error range: CP14 2) ages. Interestingly, the uppermost 430 sample of their travertine (ST4) is younger than the underlying (CP13\_1-5; 431 CP13 1-4) or guasi coeval (considering the error range: ST1; ST3) to the 432 underlying ST1 and ST3. Although problems seem to exist with the travertine 433 ages, Billi et al. (2017) did not discuss any possible source of errors. The age 434 results, irrespective of how reliable and of high precision in terms of the 435 laboratory methods, cannot be considered as a solid argument to support their 436 model for vein formation. 437

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## 439 **5. Conclusions**

Billi et al. (2017) proposed that travertine deposits on horizontal and/or 440 inclined surfaces can subsequently be folded non-tectonically by volume-441 change processes. Herein, we have shown that: (i) a comparison with active 442 travertine systems and the extensive literature on the topic do not support the 443 interpretations proposed by Billi et al. (2017); (ii) the criteria proposed by Billi 444 et al. (2017) are largely disproved, as most are based on questionable 445 interpretations; iii) the deformation model proposed by Billi et al. (2017) is not 446 supported by evidence that is available from spring systems found throughout 447 448 the world. Therefore, we do not recommend using the nine criteria that Billi et 449 al. (2017) proposed to distinguish between primary and secondary travertine structures. 450

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958 Fig. 1 – a) Terraced depositional system at Saturnia (Italy) with pools and

- rims, where calcite crystallize as a result of CO<sub>2</sub> degassing from flowing 959 carbonate-rich thermal waters; b) example of crystalline layers 960 (crystalline crusts) deposited on the slope system and formed by dendritic 961 calcite crystals; c) detail of the crystalline crusts indicated in (b). d-e) 962 Example of a slope microterraced deposit formed by dendritic calcite 963 crystals forming crystalline crusts (the so-called "chimney-like veins" by 964 Billi et al., 2017); e-f) details of the inset in (e): note the different shape 965 of the crystal-fans along the same level, emphasizing that that the 966 direction of the calcite crystal growth cannot be based on the fan-like 967 968 crystal arrangements as proposed by Billi et al. (2017).
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Fig. 2 - Present-day depositional systems and related macro-facies. a) 971 Terraced slope depositional system at Pamukkale (Turkey); b) Terraced 972 slope depositional system at Karahayıt (Turkey); note the stepped 973 morphology of the terraced slope with metre-scale pools separated by 974 975 round rims at the pool margin and vertical walls. c-d) Detail of a pool 976 (indicated in b) illustrating the site of precipitation of travertine with different fabrics, such as shrubs, radial pisoids and coated gas bubbles (cf 977 Guo and Riding, 1998); e) micro-terraced slope system; f) detail of micro-978 979 terraces showing pools and rims, where the rims are built by crystalline dendrites and pools are sites of precipitation of different travertine fabrics 980 such as shrubs and porous travertine (e.g. coated gas bubbles). 981

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- Fig. 3 Examples of banded calcite veins filling cracks that cut across layers or 983 follow bedding surfaces; a) banded calcite vein crossing late Pleistocene 984 travertine layers at Bagno Vignoni (southern Tuscany, Italy); b) banded 985 calcite veins system crossing a Pleistocene fissure ridge-type travertine 986 deposit (Akköy fissure-ridge) in the Denizli Basin (Turkey; c) banded 987 calcite vein filling a sub-vertical fracture in the wall of the Akköy fissure 988 ridge from Denizli Basin (Turkey); d) detail of the inset indicated in (b); e) 989 banded calcite veins system filling sub-vertical fractures crossing the 990 991 middle Pleistocene fissure ridge-type travertine deposit (Çukurbağ fissureridge) in the Denizli Basin (Turkey; f) detail of the inset indicated in (e); 992 993 g) sub-horizontal banded calcite vein filling a fracture sub-parallel to the bedding surfaces in the late Pleistocene-Holocene travertine deposits at 994 995 Cava Campo Muri (Rapolano Terme, Italy); h) Sub-horizontal and lowangle banded calcite veins system filling a fracture that cut across layers 996 997 or follow bedding surfaces in the late-Pleistocene-Holocene travertine deposits at Cava Campo Muri (Rapolano Terme, Italy); i) detail of the 998 999 inset indicated in (h). 1000

- Fig. 4 Photomicrographs of microfabrics of crystalline crusts and banded 1001 calcite veins filling fractures cutting across travertine beds. a-b) example 1002 of crystalline crust from Rapolano Terme (Italy); c-d) example of banded 1003 1004 calcite vein from the Denizli Basin (Turkey). Please note fabric, dimension and crystal morphology that are different from the interpreted veins by 1005 Billi et al. (2017). Many other examples of micro-fabrics are illustrated in 1006 numerous publications (Gandin and Capezzuoli, 2014; Della Porta, 2015; 1007 Croci et al., 2016; Della Porta et al., 2017 and references therein) to 1008 1009 which the readers are addressed for more details.
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- Fig. 5 Photographs of saw-cut walls exposed in the Pianetti guarry near 1011 1012 Saturnia (southern Tuscany, Italy), the same outcrops reported in Billi et al. 2017. a-b) Slope deposit formed by crystalline crusts giving rise to 1013 terraces in a prograding pools system (the so-called "chimney-like veins" 1014 by Billi et al. 2017, enlarged in c) and the final slope accumulation. The 1015 slope deposit is unconformably overlain by subhorizontal porous strata 1016 formed by shrub facies (typical of subhorizontal pools) terminating in 1017 onlap against the slope depositional profile. c) Decimeter-scale pools and 1018 rims characterized by prograding and aggrading different fabric types. d) 1019 Cross-section of a progradational terraced slope with pools bordered by 1020 1021 round rims prograding and aggrading. The terraced system is unconformably overlain by subhorizontal strata onlapping against the pool 1022 rims: this geometric configuration is a primary depositional feature and 1023 should not be explained as a secondary features (i.e. the result of syn-1024 diagenetic folds caused by laterally-confined volume expansion through 1025 hydrothermal incremental veining) as proposed by Billi et al. (2017) 1026 because the subhorizontal porous strata in onlap do not appear deformed 1027 by the alleged syn-diagenetic folding. 1028 1029
- 1030 Fig. 6 – a) Irregular geometrical setting of travertine beds visible in saw-cut 1031 walls exposed in the Pianetti quarry near Saturnia (southern Tuscany, Italy), and interpreted by Billi et al. (2017) as the result of refold 1032 1033 structures (cf their Fig. S10); b) Example of an active cascade travertine 1034 depositional system at Bagni San Filippo (southern Tuscany, Italy), where 1035 the sub-vertical slopes explain the geometrical setting of the travertine beds illustrated in (a) and interpreted by Billi et al. (2017) as secondary 1036 1037 fold structures (i.e. refolded structures). c) Cross-section of a terraced slope; d) saw-tooth shape of travertine layers deriving from an orthogonal 1038 1039 cross-section of the terraced slope: the saw-tooth shape is the result of the progradational growth of the microterraced slope. 1040



Fig. 1 – a) Terraced depositional system at Saturnia (Italy) with pools and rims, where calcite crystallize as a result of CO2 degassing from flowing carbonate-rich thermal waters; b) example of crystalline layers (crystalline crusts) deposited on the slope system and formed by dendritic calcite crystals; c) detail of the crystalline crusts indicated in (b). d-e) Example of a slope microterraced deposit formed by dendritic calcite crystals forming crystalline crusts (the so-called "chimney-like veins" by Billi et al., 2017); e-f) details of the inset in (e): note the different shape of the crystal-fans along the same level reinforcing the fact that the direction of the calcite crystal growth cannot be based on the fans-like crystal arrangement as proposed by Billi et al. (2017). Figure 2 Click here to download high resolution image



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#### Figure 3 Click here to download high resolution image



Fig. 3 - Examples of banded calcite veins filling cracks that cut across layers or follow bedding surfaces; a) banded calcite veins system crossing late Pleistocene travertine layers at Bagno Vignoni (southern Tuscany, Italy); b) banded calcite veins system crossing a Pleistocene fissure ridge-type travertine deposit (Akköy fissure-ridge) in the Denizli Basin, southwestern Turkey; c) banded calcite vein filling a sub-vertical fracture in the wall of the Akköy fissure ridge (Denizli Basin, southwestern Turkey); d) detail of the inset indicated in (b); e) banded calcite veins system filling sub-vertical fractures crossing the middle Pleistocene fissure ridge-type travertine deposit (Cukurbağ fissure-ridge) in the Denizli Basin, southwestern Turkey; f) detail of the inset indicated in (e); g) sub-horizontal banded calcite vein filling a fracture sub-parallel to the bedding surfaces in the late Pleistocene-Holocene travertine deposits at Cava Campo Muri (Rapolano Terme, Italy); h) Sub-horizontal and low-angle banded calcite veins system filling a fracture that cut across layers or follow bedding surfaces in the Late-Pleistocene-Holocene travertine deposits at Cava Campo Muri (Rapolano Terme, Italy); i) detail of the inset indicated in (h).



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