

# 1 The permeability of stylolite-bearing limestone

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## 14 **Abstract**

15       Stylolites are planar features that form due to intergranular pressure solution. Due to their  
16 planar geometry and relative abundance in limestone reservoirs, their impact on regional fluid flow  
17 has attracted considerable interest. We present laboratory permeability data that show that  
18 stylolites can be considered as conduits for flow in the stylolite-bearing limestones measured. A  
19 combination of analysis techniques shows that this is due to a zone that surrounds these stylolites  
20 that is more porous and contains larger pores than the host rock. Our data also show that the water  
21 permeability of a sample containing a stylolite parallel to fluid flow is typically lower than its  
22 permeability to gas, explained here as a result of the expansion of minor amounts of clay found in  
23 the stylolite, and that, due to their microstructural similarities, tectonic and sedimentary stylolites  
24 affect sample permeability similarly. Finally, we show that the permeability anisotropy that

25 develops in the rock mass due to the presence of sedimentary stylolites makes it appear as though  
26 the stylolites are acting as barriers to fluid flow, and may explain the discrepancy between  
27 laboratory measurements and field-scale observations. This approach can provide estimates for the  
28 equivalent permeability, and permeability anisotropy, for stylolite-bearing limestone reservoirs  
29 worldwide.

30

31 **Keywords:** Stylolite; limestone; permeability; synchrotron X-ray computed tomography; scanning  
32 electron microscopy

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34 **Highlights**

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- 36 • Stylolites in limestones are conduits for flow, not barriers to flow.
- 37 • Stylolites are characterised by zone of higher porosity than the host rock.
- 38 • The high-porosity stylolite zone contains larger pores than in the host rock.
- 39 • Pores within stylolites are less spherical: stylolites create the high-porosity zone during  
40 their formation.
- 41 • Stylolites create a permeability anisotropy that may make them falsely appear as barriers to  
42 flow.

## 43 **1 Introduction**

44           Stylolites are planes of insoluble minerals that form in rocks as soluble minerals are  
45 removed by pressure solution (e.g., Park and Schot, 1968; Wanless, 1979; Nenna and Aydin, 2011;  
46 Croizé et al., 2013; Toussaint et al., 2018). They are common in limestones due to the relatively high  
47 solubility of calcite (e.g., Tondi et al., 2006; Fabricius and Borre, 2007; Benedicto and Schultz, 2010;  
48 Smith et al., 2011; Rustichelli et al., 2012; Agosta et al., 2012; Laronne Ben-Itzhak et al., 2014;  
49 Rustichelli et al., 2015; Martín-Martín et al., 2018), but are also found in sandstones (Heald, 1955;  
50 Walderhaug, 1996; Bjørkum et al., 1998; Walderhaug and Bjørkum, 2003; Emmanuel et al., 2010).  
51 Stylolites form perpendicular to the major compressive stress and are commonly found sub-parallel  
52 to bedding (formed by overburden stresses; “sedimentary stylolites”), but can form sub-  
53 perpendicular to bedding due to tectonic stresses (“tectonic stylolites”; e.g., Railsback and Andrews,  
54 1995; Ebner et al., 2010a). Although macroscopically planar, stylolites are morphologically variable  
55 on the meso- and microscale (e.g., Karcz and Scholz, 2003; Renard et al., 2004; Schmittbuhl et al.,  
56 2004; Rolland et al., 2012; Laronne Ben-Itzhak et al., 2012; Rolland et al., 2014; Koehn et al., 2016).  
57 Their roughness is thought to be a function of the magnitude of the stress under which they formed  
58 (e.g., Koehn et al., 2007; Ebner et al., 2009a; Koehn et al., 2012), the heterogeneity of the host  
59 material (e.g., Andrews and Railsback, 1997; Brouste et al., 2007; Ebner et al., 2009b, 2010b; Koehn  
60 et al., 2012), and/or the competition between long-range elastic redistribution and surface tension  
61 forces along the interface (e.g., Schmittbuhl et al., 2004; Renard et al., 2004).

62           Due to their macroscopically planar form, the influence of stylolites on fluid flow and  
63 reservoir compartmentalisation has drawn considerable interest. A handful of experimental studies  
64 have shown that stylolites can provide conduits for flow (Lind et al., 1994; Mallon and Swarbrick,  
65 1998; Heap et al., 2014; Rustichelli et al., 2015), challenging paradigms that stylolites present  
66 barriers to fluid flow (Dunnington, 1967; Nelson, 1981; Burgess and Peter, 1985; Koepnick, 1987;  
67 Finkel and Wilkinson, 1990; Dutton and Willis, 1998; Alsharhan and Sadd, 2000). Heap et al.

68 (2014), for example, measured the permeability of limestone samples that contained no stylolites,  
69 one stylolite perpendicular to the imposed fluid flow, or one stylolite parallel to flow. They found  
70 that samples containing stylolites parallel to flow were about an order of magnitude more  
71 permeable than the stylolite-free material. They concluded that this was likely due to a zone of  
72 enhanced porosity surrounding the stylolite, a conclusion supported by microstructural  
73 observations (Carozzi and von Bergen, 1987; Raynaud and Carrio-Schaffhauser, 1992; van Geet et  
74 al., 2000; Gringas et al., 2002; Padmanabhan et al., 2015). The higher porosity zone surrounding a  
75 stylolite has also been shown to reduce the uniaxial compressive strength of a stylolite-bearing  
76 sample (Baud et al., 2016). However, although stylolites themselves may act as conduits for fluid  
77 flow (e.g., Heap et al., 2014; Rustichelli et al., 2015), we highlight that they are the by-product of a  
78 process whereby dissolved materials are often precipitated into the pore space of the adjacent rock,  
79 thereby lowering the porosity, and presumably permeability, relative to the original host rock (see  
80 Toussaint et al. (2018) for a review). Therefore, formations containing abundant stylolites will  
81 likely be characterised by lower porosities and permeabilities than neighbouring stylolite-free  
82 formations. Indeed, stylolite density has been measured to be inversely proportional to porosity in  
83 some limestone formations (e.g., Alsharhan and Sadd, 2000).

84 We extend the study of Heap et al. (2014) by providing new porosity-permeability data for  
85 stylolite-bearing limestones. We also (1) compare the gas and water permeability of stylolite-  
86 bearing limestones and (2) compare the permeabilities of limestone samples containing  
87 sedimentary and tectonic stylolites. Our experimental data are supported by microstructural  
88 observations (scanning electron microscope, SEM), multi-resolution (voxel size of 6.27 and 0.7  $\mu\text{m}$ )  
89 synchrotron X-ray computed tomography (CT), and estimations of the average pore radius of the  
90 flow path used by gas particles determined using the Klinkenberg slip factor. Finally, and using our  
91 experimental data, we consider the “upscaled” permeability of a limestone rock mass containing  
92 stylolites.

93

94 **2 Experimental materials and methods**

95           We selected six stylolite-bearing limestones for this study: two from open quarries in  
96 Burgundy (France) (Corton limestone and Comblanchien limestone, both Jurassic) and four from  
97 cores drilled around the ANDRA Underground Research Laboratory near Bure (France) (one from  
98 the Middle Jurassic “Dogger” series and three from the Late Jurassic Oxfordian stage). The  
99 porosities and gas permeabilities of some of the samples from Bure were previously presented in  
100 Heap et al. (2014). We provide here new water permeability data on these samples and new  
101 porosity and permeability (gas and water) data for additional samples taken from one of the cores  
102 from Bure (from the Late Jurassic Oxfordian stage) and the samples from Burgundy.

103           We first quantified the mineral content of our experimental materials using X-ray powder  
104 diffraction (XRPD). Powdered samples of each of the limestones were ground for 10 minutes in  
105 alcohol using an agate pestle and mortar. The XRPD analyses were performed on powdered mounts  
106 (using nominally zero-background Si sample holders and 10-20 mg of material) using a Bruker D-  
107 5005  $\theta$ - $2\theta$  Bragg-Brentano diffractometer equipped with Ni-filtered  $\text{CuK}\alpha$  radiation. The obtained  
108 XRPD patterns were first checked for their crystalline content using search-match comparisons  
109 with XRPD standards contained in the inorganic crystal structure database (ICSD). The XRPD  
110 patterns with more than one crystalline phase were then refined using the software EXPGUI-GSAS  
111 (Larson and Von Dreele 1994; Toby 2001). EXPGUI-GSAS uses the Rietveld method to derive  
112 crystallographic parameters and phase abundances (wt. %). More detailed descriptions of the  
113 Rietveld refinement method are reported in Iezzi et al. (2004; 2010 and references therein). XRPD  
114 analysis was performed on (1) stylolite-free material and (2) on samples cut to contain a stylolite,  
115 but with as little host rock as possible (in an attempt to identify the minerals forming the stylolite).  
116 In addition, minerals within the stylolites were also identified using energy-dispersive X-ray

117 spectroscopy (EDS) during our SEM analyses. The mineral content for the Bure samples was  
118 previously presented in Heap et al. (2014).

119         The first of the Oxfordian limestones (O1; depth = 159 m) is a heterogeneous allochemical  
120 limestone that contains ooids, peloids, shell fragments, and fossil foraminifera within a micrite  
121 matrix (Figure 1a). The ooids are typically 0.1-0.25 mm in diameter (Figure 1a). The peloids are  
122 noticeably larger than the ooids (Figure 1a); some peloids have diameters greater than 1 mm. Shell  
123 fragments in O1 can be many millimetres in length. O1 is predominately calcite (99 wt. %) with  
124 subordinate dolomite (<1 wt. %) (Table 1). The second Oxfordian limestone (O3; depth = 174 m) is  
125 a well-sorted allochemical limestone that contains ooids, typically 0.25-0.5 mm in diameter, within  
126 a micrite matrix (Figure 1b). O3 is predominately calcite (99 wt. %) with subordinate dolomite (<1  
127 wt. %), gypsum (<1 wt. %), and pyrite (<<1 wt. %) (Table 1). The third Oxfordian limestone (O6;  
128 depth = 364 m) is a very heterogeneous allochemical limestone that contains peloids, shell  
129 fragments (>1 mm), and fossil foraminifera (>1 mm) within a micrite matrix (Figure 1c). O6 is  
130 predominately calcite (99 wt. %) with subordinate dolomite (<1 wt. %) and pyrite (<<1 wt. %)  
131 (Table 1). The “Dogger” limestone (D3; depth 747 m) is an orthochemical limestone (micrite)  
132 (Figure 1d) composed of 93 wt. % calcite, 4 wt. % dolomite, 3 wt. % quartz, and subordinate pyrite  
133 (<<1 wt. %) (Table 1). Corton limestone is an allochemical limestone that contains peloids  
134 (typically 0.2-1 mm in diameter) within a micrite matrix (Figure 1e). Corton limestone is  
135 predominately calcite (99 wt. %) with subordinate quartz (<1 wt. %) (Table 1). Comblanchien  
136 limestone is a heterogeneous allochemical limestone that contains ooids, peloids, shell fragments,  
137 and fossil foraminifera within a micrite matrix (Figure 1f). Allochems are typically between 0.1 and  
138 1 mm in diameter (Figure 1f). Comblanchien limestone is essentially entirely calcite in composition  
139 (99 wt. %) (Table 1).

140         Examples of the stylolites in these materials are shown in Figures 2, 3, 4, and 5.  
141 Qualitatively, there are clear differences between the sedimentary stylolites in terms of thickness

142 and roughness/tortuosity (Figures 2, 3, and 4). The thickest stylolites (up to 2-3 mm) are found in  
143 the D3 samples (Figure 3a). The stylolites in Corton limestone are the most rough/tortuous (Figure  
144 4a); the least rough/tortuous stylolites are found in samples O3 (Figure 2b) and the Comblanchien  
145 limestone (Figure 4b). Stylolites found in the most heterogeneous limestone—sample 06—are  
146 correspondingly anastomosing (Figure 2c). We also provide images of tectonic and sedimentary  
147 stylolites found in sample O3 (Figure 5). There are no discernable differences between the  
148 thickness and roughness/tortuosity between the tectonic and sedimentary stylolites in sample O3  
149 (Figure 5). A combination of XRPD and EDS analyses found that the stylolites typically consist of  
150 dolomite and/or quartz, with minor quantities of pyrite and organic matter/clay (Table 1).

151         Cylindrical samples (20 mm in diameter and nominally 40 mm in length) were cored from  
152 the blocks/cores. Samples were prepared to contain (1) one stylolite perpendicular to the axis of  
153 the core (i.e. perpendicular to the imposed flow direction), (2) one stylolite parallel to the axis of  
154 the core (i.e. parallel to the imposed flow direction), or (3) no stylolite (where possible, stylolite-  
155 free samples were prepared in two or three orthogonal directions). The samples containing no  
156 stylolites were typically prepared from material 5-10 cm from the stylolite studied. We also  
157 prepared samples containing tectonic stylolites either perpendicular or parallel to the core axis  
158 from one of the Oxfordian limestones (sample O3). We note that, following sample preparation, our  
159 samples did not contain any obvious stylolite-associated fractures. Representative photographs of  
160 the 20 mm-diameter samples prepared for laboratory testing (stylolite-free, one stylolite  
161 perpendicular to the sample axis, and one stylolite parallel to the sample axis) are provided in  
162 Figures 6 and 7. Figure 8 shows photographs of samples of O3 containing tectonic and sedimentary  
163 stylolites.

164         The connected porosity of each sample was determined using the triple weight water  
165 saturation technique (Guéguen and Palciauskas, 1994). Gas (argon or nitrogen) and water (distilled  
166 water) permeabilities were then measured in a hydrostatic pressure vessel under a confining



167 pressure of 2 MPa. All measurements of water permeability were performed using the steady-state  
168 flow method. Following microstructural equilibrium, a pressure differential was imposed across the  
169 sample and the flow rate was measured using an electronic balance (with a precision  $\pm 0.0005$  g).  
170 Once steady-state flow had been established, the water permeability  $k_{water}$  was determined using  
171 Darcy's relation:

$$\frac{Q}{A} = \frac{k_{water}}{\eta L} (P_{up} - P_{down}), \quad (1)$$

173  
174 where  $Q$  is the volumetric flow rate,  $A$  is the cross-sectional area of the sample,  $P_{up}$  and  $P_{down}$   
175 represent the upstream and downstream pressure, respectively (where  $P_{down}$  is the atmospheric  
176 pressure),  $L$  is the length of the sample,  $k_{water}$  is the permeability to water, and  $\eta$  is the viscosity of  
177 the pore fluid (taken here as  $1.008 \times 10^{-3}$  Pa s). A pressure differential (i.e.  $P_{up} - P_{down}$ ) of 0.5 MPa  
178 was used for all measurements reported herein.

179 Gas (argon or nitrogen) permeability was measured using either the steady-state method  
180 (for high-permeability samples) or the pulse-decay method (for low-permeability samples). For the  
181 steady-state method, a pressure differential was imposed across the sample (following  
182 microstructural equilibrium) and the outlet flow rate was measured using a flow meter. Since the  
183 pore fluid is compressible, the raw permeability to gas  $k_{gas\_raw}$  is expressed as (Scheidegger,  
184 1974):

$$\frac{Q}{A} = \frac{k_{gas\_raw}}{\eta L} \frac{(P_{up})^2 - (P_{down})^2}{2P_{down}}, \quad (2)$$

186  
187 where  $\eta$ , the viscosity of the pore fluid, was taken as  $2.21 \times 10^{-5}$  and  $1.78 \times 10^{-5}$  Pa s for argon and  
188 nitrogen, respectively. Steady-state volumetric flow rate  $Q$  measurements were taken under several

189 pore pressure differentials (i.e.  $P_{up} - P_{down}$ , where  $P_{down}$  is the atmospheric pressure) to check  
 190 whether any auxiliary corrections were required. We first plot  $1/k_{gas\_raw}$  as a function of  $Q$  to  
 191 check whether the Forchheimer correction is required (Forchheimer, 1901). The correction is  
 192 necessary if these data can be well described by a linear fit with a positive slope. The Forchheimer-  
 193 corrected permeability is taken as the inverse of the  $y$ -intercept of the best-fit linear regression in  
 194 the plot of  $1/k_{gas\_raw}$  as a function of  $Q$ . If the Forchheimer correction is not required, we then  
 195 check whether the Klinkenberg correction is required (Klinkenberg, 1941). To do so, we plot  
 196  $k_{gas\_raw}$  as a function of the reciprocal mean pressure  $1/P_m$ , where  $P_m$  is the mean pore fluid  
 197 pressure (i.e.  $(P_{up} + P_{down})/2$ ). The Klinkenberg correction is required if these data can be well  
 198 described by a linear fit with a positive slope and, if true, the Klinkenberg-corrected permeability  
 199 can be taken as the  $y$ -intercept of the best-fit linear regression in the plot of  $k_{gas\_raw}$  as a function of  
 200  $1/P_m$ . The Klinkenberg correction was required for all samples measured using the steady-state  
 201 method; the Forchheimer correction was not required.

202 We used the pulse-decay method (Brace et al., 1968) to measure the gas permeability of the  
 203 low-permeability samples. Following microstructural equilibrium at the target confining pressure,  
 204 the decay of an initial pore pressure differential ( $P_{up} - P_{down} = 0.5$  MPa, where  $P_{down}$  is the  
 205 atmospheric pressure) was monitored using a pressure transducer following the closure of the  
 206 upstream pressure inlet. The gas permeability  $k_{gas\_raw}$  was then determined using the following  
 207 relation:

$$k_{gas\_raw} = 2 \frac{\eta L}{A} \frac{V_{up}}{P_{up}^2 - P_{down}^2} \frac{dP_{up}}{dt}, \quad (3)$$

209 where  $V_{up}$  is the volume of the upstream pore pressure circuit ( $7.8 \times 10^{-6}$  m<sup>3</sup>) and  $t$  is time. As  
 210 before, we checked whether these data required any auxiliary corrections (the Forchheimer or  
 211

212 Klinkenberg correction). The Klinkenberg correction was required for all samples measured using  
213 the pulse-decay method; the Forchheimer correction was not required. A detailed description of  
214 these permeability methods is available in Heap et al. (2017).

215

### 216 **3 Results**

217 The gas permeability data for the stylolite-free limestones as a function of connected  
218 porosity are shown in Figure 9a. The data of Lind et al. (1994), a study that also measured the  
219 permeability of stylolite-bearing carbonate rocks, are also included on Figure 9 because they  
220 preserve a higher porosity (porosity > 0.2) than the samples measured herein. Our data show that  
221 gas permeability increases as connected porosity is increased, in accordance with previously  
222 published studies on the permeability of limestones (e.g., Ehrenberg et al., 2006; Zinszner and  
223 Pellerin, 2007), and that there is no measurable permeability anisotropy in the studied materials  
224 (Figure 9a contains data on samples cored in orthogonal directions, see Table 2). The difference  
225 between permeability to gas and permeability to water in the stylolite-free samples appears to  
226 depend on the connected porosity: permeability to gas can be a factor of 4.5 higher at low porosity  
227 (porosity < 0.05) and the ratio between gas and water permeability is essentially unity at the  
228 highest tested porosity (porosity ~0.15) (Figure 10a).

229 The porosity-permeability data for the stylolite-free and stylolite-bearing (perpendicular  
230 and parallel to flow) limestones are shown in Figure 9b, together with the high-porosity (porosity >  
231 0.2) data of Lind et al. (1994). Our data show that (1) the permeabilities of the samples containing  
232 stylolites perpendicular to the direction of flow are similar to those of the stylolite-free samples and  
233 (2) the permeabilities of the samples containing stylolites parallel to the direction of flow are  
234 characterised by permeabilities higher than those of the stylolite-free samples (Figure 9b). In detail,  
235 we notice that larger differences between the permeability of the samples containing stylolites  
236 parallel to the direction of flow and the stylolite-free samples are observed at lower connected

237 porosities (Figure 9b). For example, the permeability of stylolite-bearing Corton limestone  
238 (porosity  $\sim 0.03$ ) can be two or three orders of magnitude higher than the stylolite-free material  
239 (Figure 9b). The ratio of gas to water permeability for all the samples tested (including stylolite-free  
240 samples and samples containing stylolites perpendicular and parallel to flow) is plotted as a  
241 function of connected porosity in Figure 10b. As for the stylolite-free limestones (Figure 10a), high-  
242 porosity (porosity  $\sim 0.15$ ) samples containing stylolites show little difference between gas and  
243 water permeability (Figure 10b). The gas permeabilities of the low-porosity samples containing  
244 stylolites are higher than their water permeabilities; this is especially true for the low-porosity  
245 samples containing stylolites parallel to flow (the difference for one sample is more than an order  
246 of magnitude) (Figure 10b).

247 Our data also show that there is essentially no difference between the influence of  
248 sedimentary and tectonic stylolites on the permeability of our limestone samples (Figures 9b and  
249 10b; Table 2).

250

## 251 **4 Discussion**

252

### 253 4.1 Barriers to or conduits for fluid flow?

254 Our new permeability data are in agreement with the conclusion of Heap et al. (2014) and  
255 Rustichelli et al. (2015): the stylolites measured are not barriers to flow, but conduits for flow  
256 (Figure 9b). Heap et al. (2014) postulated that a zone of higher porosity surrounds a stylolite and  
257 that it is this high-porosity zone that enhances the circulation of fluids, as suggested by Carozzi and  
258 von Bergen (1987), Raynaud and Carrio-Schaffhauser (1992), and Van Geet et al. (2000). The  
259 greater increase in permeability in the low-porosity samples (when comparing the permeability of  
260 a stylolite-free sample to a sample containing a stylolite parallel to flow) (Figure 9b) is likely a  
261 consequence of their low matrix permeabilities. Conduits for flow have a much greater impact on

262 the equivalent permeability of low-porosity samples than on high-porosity samples, since the  
263 matrix permeability of a high-porosity sample is much closer to the permeability of the fracture (as  
264 observed in variably porous fractured materials; e.g., Heap and Kennedy, 2016; Kushnir et al.,  
265 2018).

266 To image the hypothesised zone of higher porosity, we first provide a backscattered  
267 scanning electron microscope (BSE) image of a stylolite within sample D3, selected due to its low  
268 matrix porosity and permeability. This image shows that the matrix-stylolite interface is populated  
269 by numerous micropores, typically only a few microns in diameter (white arrows; Figure 11). To  
270 better resolve the porosity, and distribution of porosity, around a stylolite, we also provide multi-  
271 resolution (voxel size of 6.27 (beamline MB05) and 0.7  $\mu\text{m}$  (beamline ID19) and energy of 35 keV)  
272 three-dimensional X-ray tomography imaging performed on a stylolite within sample D3 at the  
273 European Synchrotron Radiation Facility (Grenoble, France). Because of the high-contrast between  
274 the porosity and the minerals that comprise the rock (primarily calcite, quartz, and dolomite; Table  
275 1), it is straightforward to segment the porosity so that individual pores can be imaged (Figure 12).  
276 The segmented images show that the stylolite is associated with a zone of high porosity (Figure 12),  
277 as previously measured by Baud et al. (2016). In particular, we observe that (1) the pores  
278 surrounding the stylolite are larger than those within the host rock (Figure 12) and (2) some of the  
279 pores are aligned with the teeth of the stylolite and are characterised by a “finger-like” shape  
280 (Figure 13). Indeed, analysing the X-ray tomography data (similar to X-ray tomographic analyses  
281 performed on intact porous limestones by Ji et al., 2012; 2014) show that the pores are larger  
282 inside the stylolite (Figure 14a) and that the pores within the stylolite are characterised by lower  
283 values of sphericity (where 1.0 is a perfect sphere; sphericity is defined using the Thermo Scientific  
284 Avizo toolbox as  $(\frac{1}{Shape_{VA3D}})^{1/3}$  where  $Shape_{VA3D} = Area_{3D}^3 / (36 \times \pi \times Volume_{3D}^2)$ ) (Figure 14b).  
285 The volume of an individual pore within the stylolite varies from a few  $\mu\text{m}^3$  up to  $>10^5 \mu\text{m}^3$ ; the  
286 volume of the pores outside the stylolite are all  $<10^4 \mu\text{m}^3$  (Figure 14a). The average equivalent

287 diameter of the pores inside and outside the stylolite is 36.5  $\mu\text{m}$  (standard deviation of 26.8  $\mu\text{m}$ )  
 288 and 11.1  $\mu\text{m}$  (standard deviation of 4.7  $\mu\text{m}$ ), respectively. Sphericity inside and outside the stylolite  
 289 varies from 0.2 to 0.4 and 0.4 and 0.9, respectively (Figure 14b). Since the shape of the pores are  
 290 sometimes linked to the shape of the stylolite (Figure 13), we additionally conclude that such  
 291 porosity is likely the consequence of stylolite formation (in agreement with the conclusions of  
 292 Raynaud and Carrio-Schaffhauser (1992) and Carozzi and von Bergen (1987)), rather than that  
 293 stylolites form preferentially in a zone of higher porosity (as hypothesised by Braithwaite, 1989).

294 To complement these microstructural data, we use the Klinkenberg slip factor,  $b$  (which has  
 295 the units of pressure; Table 2) (Klinkenberg, 1941), to provide an independent assessment of the  
 296 average pore radius used by the gas molecules. Since the pore radius determined using this  
 297 technique uses data from permeability experiments, it will therefore yield the average pore throat  
 298 radius (in contrast to the CT data, which provides information on the pores). Since the mean free  
 299 path is inversely proportional to  $P_m$ , Poiseuille's law for gas flow in a cylindrical tube and Darcy's  
 300 law for flow in porous media yields the following relation:

301

$$k_{gas} = k_{gas\_raw} \left( 1 + \frac{b}{P_m} \right), \quad (4)$$

302

303 where  $k_{gas}$  is the true (i.e. Klinkenberg-corrected) gas permeability (Klinkenberg, 1941). The  
 304 average pore throat radius of the flow path followed by the gas molecules,  $r$ , can be estimated using  
 305 the following relation (Civan, 2010):

306

$$r = \frac{4}{b} \eta \sqrt{\frac{\pi R_g T}{2 M_w}}, \quad (5)$$

307

308 where  $R_g$  is the ideal gas constant (taken as  $8.31 \text{ J mol}^{-1} \text{ K}^{-1}$ ),  $T$  is the temperature (taken as  $293 \text{ K}$ ),  
309 and  $M_w$  is the molar mass of the pore fluid (taken as  $0.03995$  and  $0.02802 \text{ kg mol}^{-1}$  for argon and  
310 nitrogen, respectively). The Klinkenberg slip factor has previously been used to examine the  
311 average pore throat radius of the flow path in rocks such as shales (e.g., Heller et al., 2014; Firouzi  
312 et al., 2014; Letham and Bustin, 2016) and, more recently, volcanic rocks (Heap et al., 2018) using  
313 the same, or similar, method (i.e. Equation (5)). We find that the average pore throat radius of the  
314 flow path followed by the gas molecules in the stylolite-free samples, excluding the Corton  
315 limestone samples, varies between  $\sim 0.05$  and  $0.15 \text{ }\mu\text{m}$  (Figure 15a). Excluding Corton limestone,  
316 the samples containing the highest porosities (porosity  $\sim 0.15$ ) are characterised by the largest pore  
317 throat radii ( $\sim 0.1$  to  $\sim 0.15 \text{ }\mu\text{m}$ ; Figure 15a). The average pore throat radius of the flow path  
318 followed by the gas molecules is much larger in Corton limestone, varying between  $\sim 0.2$  and  $0.35$   
319  $\mu\text{m}$  (Figure 15a). Although not obvious from our microstructural observations (Figure 1e), Corton  
320 limestone must contain larger pore throats than the other limestones measured herein. The  
321 average pore throat radii of the flow paths followed by the gas molecules in the samples containing  
322 stylolites (together with the stylolite-free samples) are provided in Figure 15b. The data show that  
323 (1) the pore throat radii forming the flow path in the samples containing stylolites perpendicular to  
324 flow are similar to the stylolite-free samples and (2) the pore throat radii along the flow path  
325 parallel to the stylolite are systematically larger than those of the other samples (Figure 15b).  
326 These data suggest that the stylolites are associated with pore throats with larger radii than those  
327 that typify the host rock. This conclusion is in agreement with our X-ray tomography analysis,  
328 which shows that the pores are larger inside the stylolite than in the host rock (Figure 14a). As  
329 expected, the radii predicted using Equation (5) are much smaller (typically  $< 1 \text{ }\mu\text{m}$ ; Figure 15) than  
330 the range of radii predicted from the X-ray tomography analysis (up to a few tens of microns). This  
331 is because the Klinkenberg analysis (Equation 5) yields the pore throat radius and the X-ray  
332 tomography analysis yields the pore radius.

333 We conclude here that stylolites present conduits for, rather than barriers to, flow (Figure  
334 9b) in limestones measured herein. This can be explained by a zone of elevated porosity (Figure 12)  
335 that contains pores and pore throats with larger radii than the host rock (Figures 14a and 15a),  
336 which we conclude must develop around a stylolite during its formation. The development of  
337 stylolitic porosity is discussed in detail in Carozzi and von Bergen (1987) and is considered the  
338 result of grainscale heterogeneities in the rock during the dissolution process.

339

#### 340 4.2 Differences between tectonic and sedimentary stylolites

341 Our permeability data suggest, for the materials studied herein, that there is essentially no  
342 difference between the influence of sedimentary and tectonic stylolites on the permeability of a  
343 stylolite-bearing sample: both sedimentary and tectonic stylolites are conduits for fluid flow  
344 (Figure 9b; Table 2). This is perhaps not surprising since they are very similar on the microscale  
345 (Figure 5) and on the sample lengthscale (Figure 8). The fact that tectonic stylolites are also  
346 conduits for flow (Table 2) further supports the hypothesis that stylolites create a zone of higher  
347 porosity during their formation (e.g., Raynaud and Carrio-Schaffhauser, 1992; Carozzi and von  
348 Bergen, 1987), rather than that they form preferentially in higher porosity layers (e.g., Braithwaite,  
349 1989).

350

#### 351 4.3 Differences between gas and water permeability

352 Differences between permeability to gas and water are typically observed in the presence of  
353 swelling clays (e.g., Faulkner and Rutter, 2000, 2003; Tanikawa and Shimamoto 2006; Davy et al.,  
354 2007; Tanikawa and Shimamoto 2009; Behnsen and Faulkner, 2011). Our XRPD analyses highlight  
355 that clays are below the detection limit in the stylolite-free material (Table 1). It is therefore  
356 perhaps surprising that we see about a fourfold difference between gas and water permeability in  
357 the low-porosity limestones (Figure 10a). A recent study found that the permeability to gas was



358 higher than permeability to water in two volcanic rocks (basalt and andesite) by a factor of up to  
359 five (Heap et al., 2018). In the absence of significant physicochemical reactions, these authors  
360 suggested that the difference in gas and water permeabilities is likely due to water adsorption on  
361 the surface of thin microstructural elements. For the stylolite-free limestones, we find that there is  
362 essentially no difference between the gas and water permeabilities for the samples characterised by  
363 the largest average pore throat radii ( $\sim 0.1$  to  $\sim 0.15$   $\mu\text{m}$ ) (Table 2), as determined using the  
364 Klinkenberg slip factor. Samples with average pore throat radii between  $\sim 0.05$  and  $0.1$   $\mu\text{m}$  are more  
365 permeable to gas than to water (Table 2). Similar to the conclusions drawn by Heap et al. (2018),  
366 we conclude here that, in the absence of clay within the intact materials (Table 1), the difference in  
367 gas and water permeabilities is likely due to water adsorption on the surface of thin ( $\sim 0.05$  to  $0.1$   
368  $\mu\text{m}$ ) microstructural elements.

369         Measurements of gas and water permeability on the samples containing stylolites show that  
370 samples containing stylolites parallel to flow are often more permeable to gas than water, by up to  
371 one order of magnitude (Figure 10b). Since average pore throat radius of the flow path followed by  
372 the gas molecules is relatively high for these samples (up to  $\sim 1$   $\mu\text{m}$ ; Figure 15b), we conclude that  
373 the difference in gas and water permeabilities in these samples are due to minor quantities of clay  
374 found within the stylolite (identified by EDS during our SEM analyses; Table 1). The expansion of  
375 clay minerals in contact with water constricts pore throats and thus reduces permeability (e.g.,  
376 Faulkner and Rutter, 2003).

377

#### 378 4.4 Implications for fluid flow in limestone reservoirs

379         Limestone forms an important component of the Earth's continental crust (Ehrenberg et al.,  
380 2006; Ford and Williams, 2013) and, as a result, the permeability of limestone reservoirs is not only  
381 important for fluid flow and pore pressure distribution within the crust, but also for the  
382 exploitation of hydrocarbon reserves.

383 Our study shows that stylolites in limestone present conduits for flow (Figure 9b) due to a  
 384 zone of elevated porosity, containing pores with larger radii than the host rock, which develops  
 385 around a stylolite during its formation (Figures 11, 12, 14, and 15). In order to consider fluid flow in  
 386 stylolite-bearing limestone reservoirs, we must first upscale our laboratory measurements. One  
 387 method to upscale such laboratory data is to first extract the permeability of a stylolite. The  
 388 permeability of a stylolite,  $k_{stylolite}$ , can be determined using a two-dimensional model that considers  
 389 flow in parallel layers (the same model used to determine the permeability of compaction bands in  
 390 Vajdova et al. (2004) and fractures in Heap and Kennedy (2016), Farquharson et al. (2016), and  
 391 Kushnir et al. (2018)):

$$k_{stylolite} = \frac{(A \cdot k_e) - (A_{intact} \cdot k_0)}{A_{stylolite}} \quad (6)$$

393  
 394 where  $A$  is the cross-sectional area of the sample,  $k_e$  is the equivalent permeability (the  
 395 permeability of the stylolite-bearing sample),  $A_{intact}$  is the area of stylolite-free material,  $k_0$  is the  
 396 stylolite-free permeability, and  $A_{stylolite}$  is the area of the stylolite. For the purpose of this exercise, we  
 397 will consider a core of Dogger limestone (D3) taken from the ANDRA Underground Research  
 398 Laboratory at Bure (Figure 16). To calculate  $k_{stylolite}$  we use the permeability of the stylolite-free  
 399 sample of D3 ( $k_0 = 3.69 \times 10^{-19} \text{ m}^2$ ; Table 2). The equivalent permeability,  $k_e$ , is permeability of the  
 400 D3 sample containing a stylolite parallel to the direction of flow ( $k_e = 5.98 \times 10^{-18} \text{ m}^2$ ; Table 2), and  
 401 we use a stylolite thickness of 1 mm (a reasonable approximation of the thickness of the stylolite in  
 402 this sample; Figure 6d). Using these values, Equation (6) yields a stylolite permeability,  $k_{stylolite}$ , of  
 403  $8.85 \times 10^{-17} \text{ m}^2$ . We can now model the equivalent permeability of a rock mass populated with  
 404 stylolites using our value for  $k_{stylolite}$  and the following relation:

405

$$k_e = \frac{(w_{intact} \cdot k_0) + (w_{stylo} \cdot k_{stylo})}{W}, \quad (7)$$

406

407 where  $w_{intact}$  is the total width of the intact material,  $w_{stylo}$  is the total width of the stylolites, and  
 408  $W$  is the length of rock considered ( $W = w_{intact} + w_{stylo}$ ). The Dogger limestone core sample  
 409 shows that there are five sedimentary stylolites over a length of about 25 cm (i.e. a stylolite density  
 410 of  $20 \text{ m}^{-1}$ ) (Figure 16). Although stylolite thickness varies (Figure 16) we will, for simplicity, assume  
 411 that the thickness of each stylolite is 1 mm. Therefore, according to our model (Equation (7));  $W =$   
 412  $250 \text{ mm}$ ;  $w_{stylo} = 5 \text{ mm}$ ;  $w_{intact} = 245 \text{ mm}$ ;  $k_0 = 3.69 \times 10^{-19} \text{ m}^2$ ;  $k_{stylo} = 8.85 \times 10^{-17} \text{ m}^2$ ), the  
 413 equivalent permeability parallel and perpendicular to bedding for the limestone core shown in  
 414 Figure 16 is  $1.80 \times 10^{-17}$  and  $3.69 \times 10^{-19} \text{ m}^2$ , respectively. In other words, the 25 cm-long sample is  
 415 50 times more permeable parallel to bedding than perpendicular to bedding. Therefore, although  
 416 stylolites are conduits for flow, they can create a permeability anisotropy in a rock unit or reservoir  
 417 that may make it appear that they form barriers to fluid flow (because permeability perpendicular  
 418 to bedding is lower than the permeability parallel to bedding) and may therefore explain the  
 419 discrepancy between laboratory measurements (that suggest that stylolites are conduits) and field-  
 420 scale investigations (that suggest that stylolites are barriers). It is important to highlight that these  
 421 equivalent permeability estimates for a stylolite-bearing rock mass are just one snapshot in the  
 422 porosity-permeability evolution of this limestone formation. For example, it is likely that, prior to  
 423 pressure-solution and the formation of the stylolites, the host rock was much more porous and  
 424 more permeable. Although not considered in our simple model, we note that the presence of  
 425 stylolites perpendicular to bedding (i.e. tectonic stylolites) will reduce such permeability  
 426 anisotropy. Indeed, if the number of tectonic stylolites equals the number of sedimentary stylolites,  
 427 no permeability anisotropy will be observed. Although tectonic stylolites are not uncommon in  
 428 limestone reservoirs (Railsback and Andrews, 1995; Ebner et al., 2010a; Figure 8), it is difficult to

429 assess their density at the ANDRA Underground Research Laboratory at Bure due to the drilling  
430 direction (perpendicular to bedding). The simple method presented above can be easily adapted to  
431 provide estimates for the equivalent permeability, and permeability anisotropy, for stylolite-  
432 bearing (both sedimentary and tectonic) limestone reservoirs worldwide.

433         Although we conclude here that our stylolites form conduits for fluid flow, we cannot rule  
434 out that some stylolites, different to those measured here, may provide barriers to flow. For  
435 example, (1) stylolites may provide barriers to flow if they are characterised by thick and  
436 continuous layers of clay-rich material, (2) an abstract by Corwin et al. (1997) suggests that  
437 stylolites associated with a cemented zone could be of lower permeability than the surrounding  
438 host rock, and (3) the modelling of Koehn et al. (2016) suggests that stylolites with simple  
439 geometries (e.g., “simple wave-like type”) may be more likely to provide barriers to flow. We also  
440 highlight that, due to differences in mineral composition and microstructure, the influence of  
441 stylolites on the permeability of sandstone may differ from their influence on the permeability of  
442 carbonate rocks (e.g., Walderhaug and Bjørkum, 2003; Emmanuel et al., 2010).

443         Therefore, and although we provide laboratory measurements for the permeability of  
444 stylolite-bearing limestone from six formations collected from two locations within France, more  
445 laboratory measurements on stylolites that are characterised by thick and continuous layers of  
446 clay-rich material are now required to further explore the role of stylolites on the regional-scale  
447 permeability of limestone reservoirs (as concluded by Bruna et al., 2018). Laboratory  
448 measurements on stylolite-bearing sandstones also offer an interesting avenue for future research.

449

## 450 **5 Conclusions**

451         The salient conclusions of this study can be summarised thusly:

452         (1) The stylolites measured herein are conduits for fluid flow, not barriers to fluid flow.

- 453 (2) The permeability of a stylolite-bearing sample is lower when measured with water than  
454 with gas. We interpret this here as the result of the expansion of minor quantities of clay  
455 found within the stylolite. The expansion of clay minerals constricts pore throats and thus  
456 reduces permeability.
- 457 (3) Sedimentary and tectonic stylolites affect sample permeability similarly. We interpret this  
458 as a result of their similar microstructures.
- 459 (4) X-ray tomography data show that the stylolites are surrounded by a zone of higher porosity  
460 that is characterised by pores larger than those found in the intact material. This explains  
461 why the stylolites measured herein are conduits for fluid flow.
- 462 (5) The presence of larger pores within the stylolite zone is supported by an analysis of the  
463 Klinkenberg slip factor, which highlights that the average pore throat radius of the flow  
464 path followed is larger when the sample contains a stylolite parallel to flow.
- 465 (6) X-ray tomography data show that the pores within the stylolite are much less spherical than  
466 those of the host rock and that they are sometimes aligned with the teeth of the stylolite.  
467 Since the shape of the pores are linked to the shape of the stylolite, we conclude that such  
468 porosity is likely the consequence of stylolite formation, rather than that stylolites form  
469 preferentially in a zone of higher porosity.
- 470 (7) Upscaling our laboratory measurements using a simple two-dimensional model that  
471 considers flow in parallel layers shows that the equivalent permeability of a stylolite-  
472 bearing limestone rock mass is higher parallel to bedding than perpendicular to bedding.
- 473 (8) The permeability anisotropy that develops in the rock mass due to the presence of stylolites  
474 makes it appear as though the stylolites are acting as barriers to fluid flow (since  
475 permeability perpendicular to bedding is lower than the permeability parallel to bedding)  
476 and may explain the discrepancy between laboratory measurements and field-scale  
477 observations.

478

479 **Acknowledgements**

480           This research was partly funded by CNRS and ANDRA (FORPRO program) and the  
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482 Rolland, Silvio Mollo, Gilles Morvan, and Bertrand Renaudié. We thank Gilles Jouillerot (Rocamat  
483 Pierre Naturelle) for his help selecting the materials at Comblanchien and Elodie Boller for her help  
484 at the European Synchrotron Radiation Facility (beamline ID19). The comments of Einat Aharonov  
485 and one anonymous reviewer helped improve this manuscript.

486

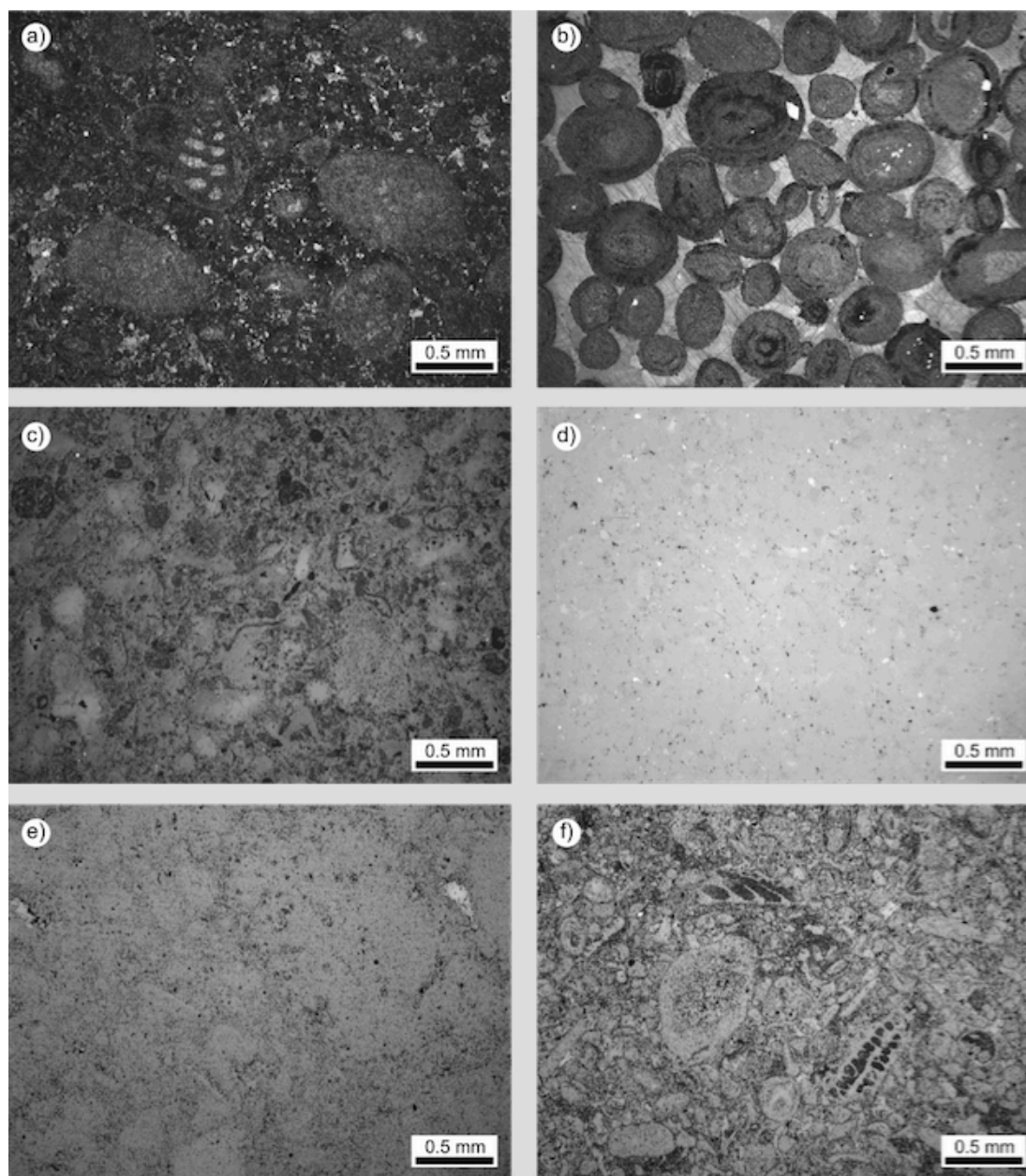
487 **Data availability**

488           Most of the data used in this study are available in Tables 1 and 2. The X-ray tomography  
489 data may be made available on request (to François Renard).

490 **Figure captions**

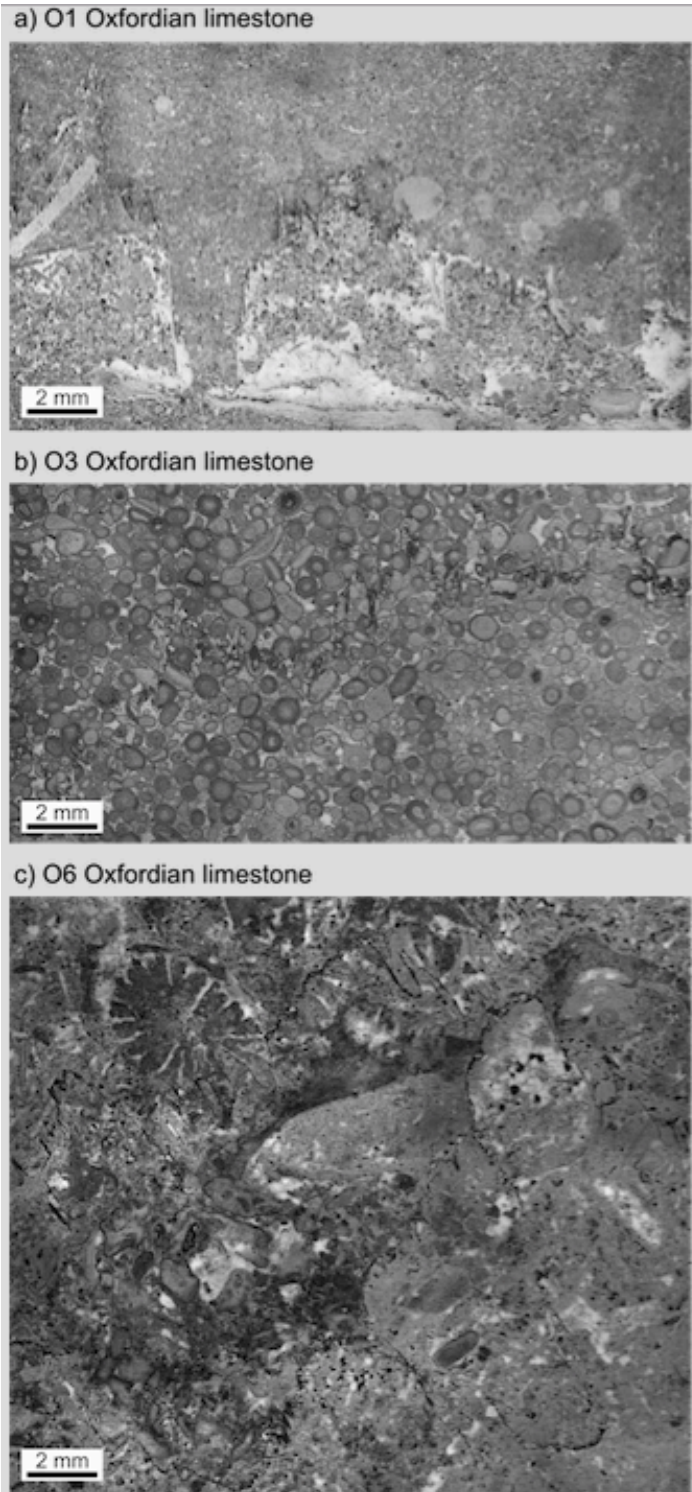
491

492 **Figure 1.** Optical microscope images of the stylolite-free (host rock) material for the studied  
493 limestones. (a) Sample O1 – Oxfordian limestone from Bure. (b) Sample O3 – Oxfordian limestone  
494 from Bure. (c) Sample O6 – Oxfordian limestone from Bure. (d) Sample D3 – “Dogger” limestone  
495 from Bure. (e) Corton limestone from Burgundy. (f) Comblanchien limestone from Burgundy.



496

497 **Figure 2.** Optical microscope images of the stylolites found within studied limestones. (a) Sample  
498 O1 – Oxfordian limestone from Bure. (b) Sample O3 – Oxfordian limestone from Bure. (c) Sample  
499 O6 – Oxfordian limestone from Bure.



500

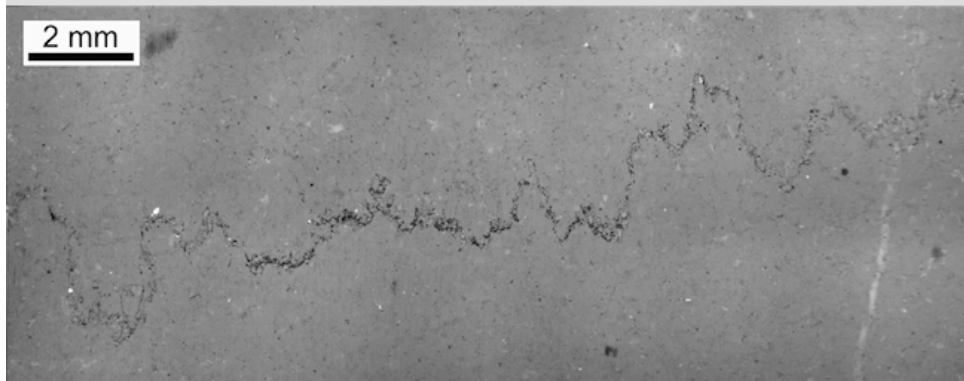


501 **Figure 3.** Optical microscope images of the stylolites found within studied limestones. (a) Sample  
502 06 - Oxfordian limestone from Bure. (b) Sample D3 - "Dogger" limestone from Bure.

a) D3 "Dogger" limestone - thick stylolite



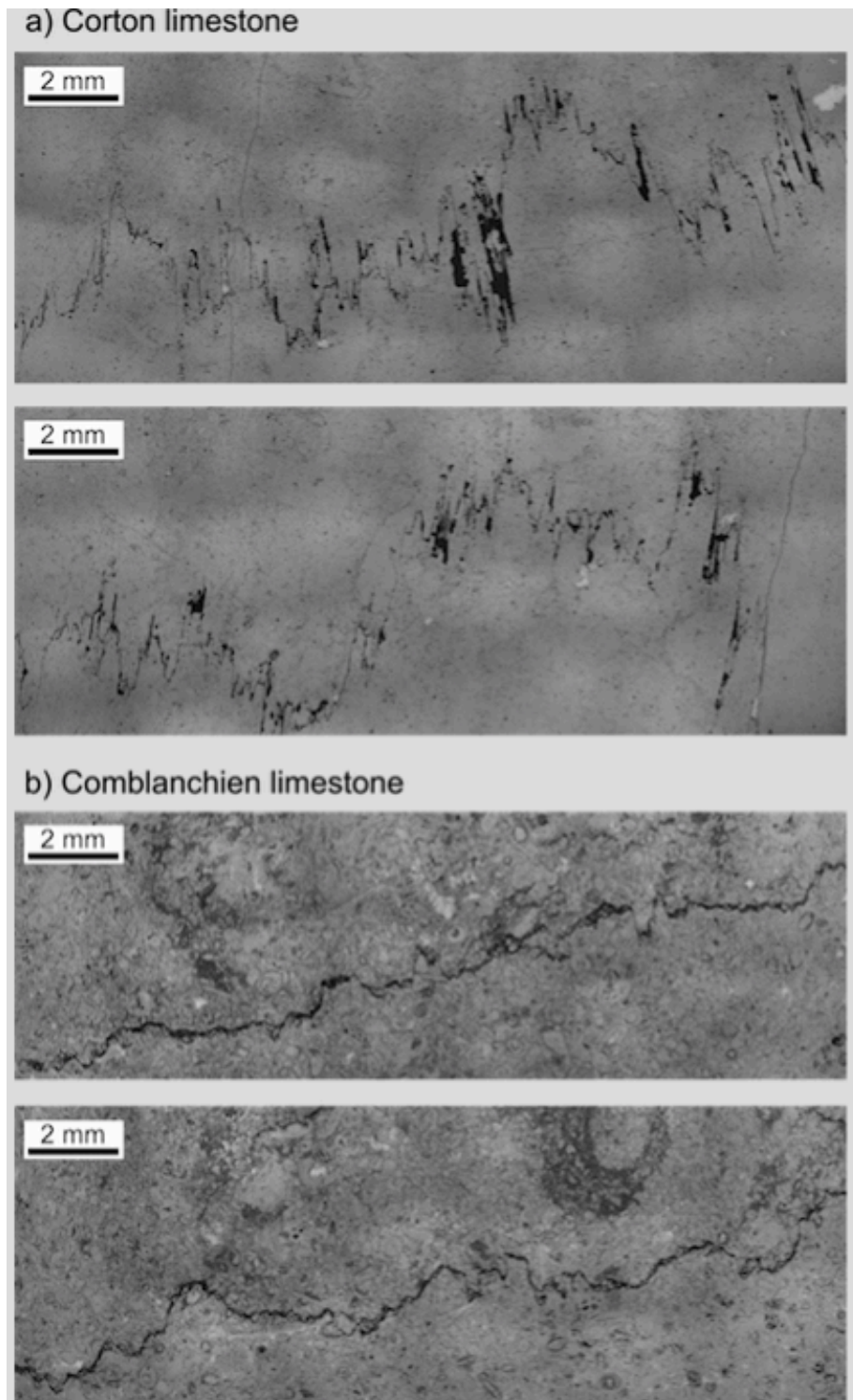
b) D3 "Dogger" limestone - thin stylolite



503

504

505 **Figure 4.** Optical microscope images of the stylolites found within studied limestones. (a) Corton  
506 limestone from Burgundy. (b) Comblanchien limestone from Burgundy.

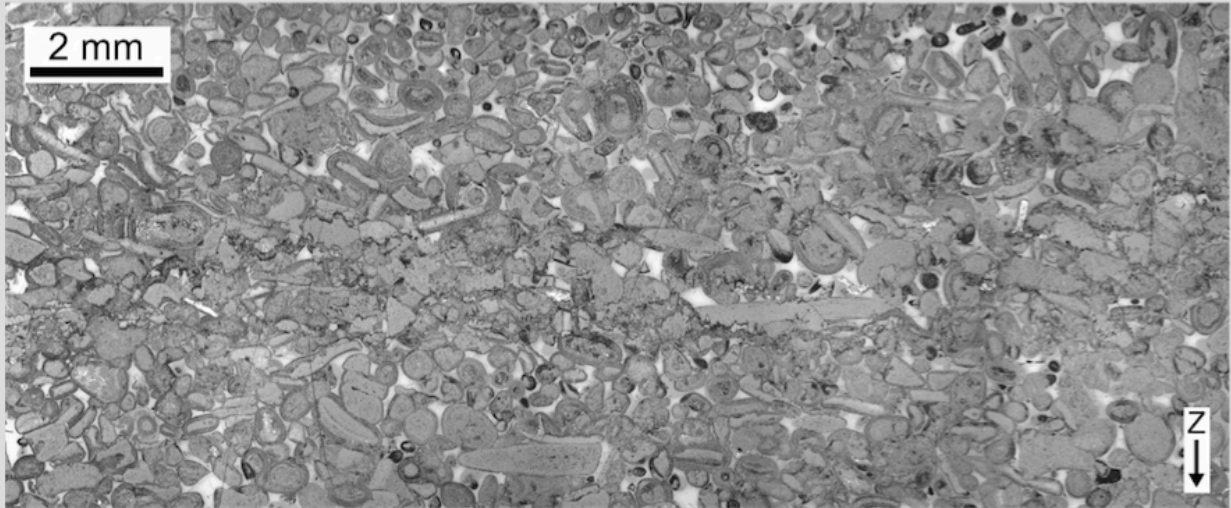


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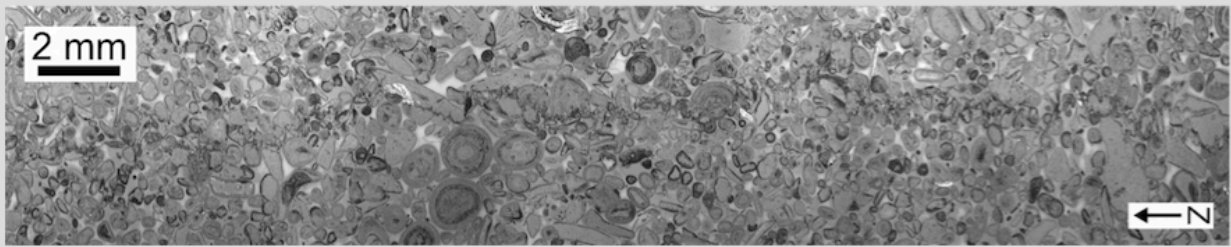
508 **Figure 5.** Optical microscope images of (a) a sedimentary stylolite and (b) a tectonic stylolite in  
509 sample O3 – Oxfordian limestone from Bure.

510

a) O3 Oxfordian - sedimentary stylolite



b) O3 Oxfordian - tectonic stylolite



511

512

513

514 **Figure 6.** Photographs of the cylindrical samples prepared for laboratory measurements. Three  
515 representative samples are shown for each lithology: an intact sample (on the left), a sample with a  
516 stylolite perpendicular to flow (in the middle), and a sample with a stylolite parallel to flow (on the  
517 right). (a) Sample O1 – Oxfordian limestone from Bure. (b) Sample O3 – Oxfordian limestone from  
518 Bure. (c) Sample O6 – Oxfordian limestone from Bure. (d) Sample D3 – “Dogger” limestone from  
519 Bure.



520

521 **Figure 7.** Photographs of the cylindrical samples prepared for laboratory measurements. Three  
522 representative samples are shown for each lithology: an intact sample (on the left), a sample with a  
523 stylolite perpendicular to flow (in the middle), and a sample with a stylolite parallel to flow (on the  
524 right). (a) Corton limestone from Burgundy. (b) Comblanchien limestone from Burgundy.

525

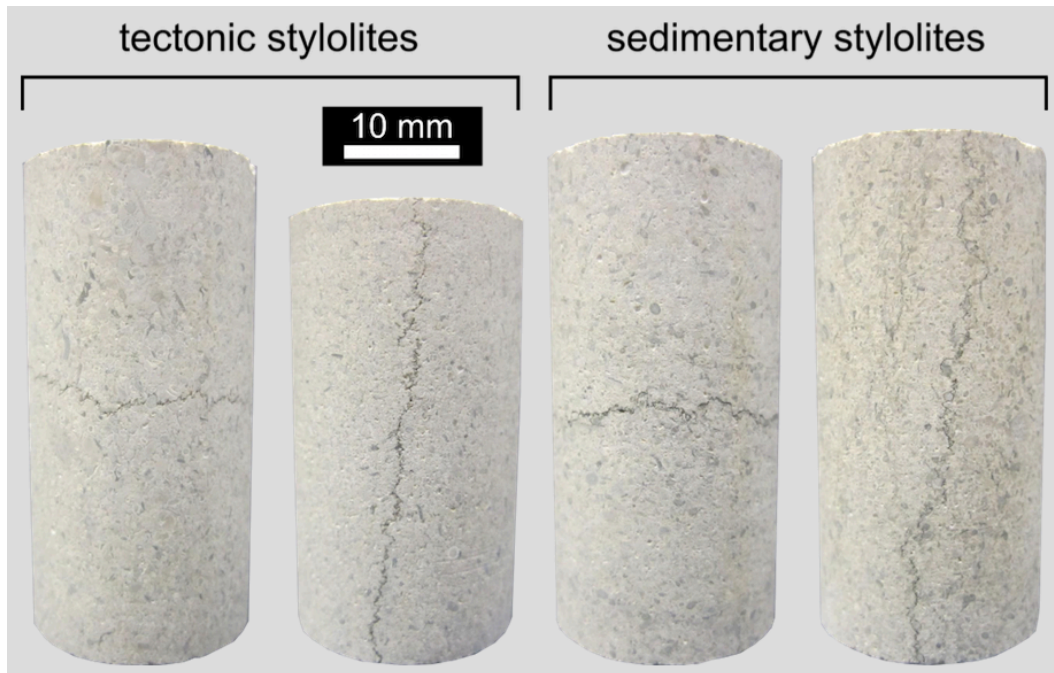


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527

528 **Figure 8.** Photographs of the cylindrical samples containing either a tectonic stylolite (the two  
529 samples on the left) or a sedimentary stylolite (the two samples on the right). All samples are from  
530 sample O3 – Oxfordian limestone from Bure.

531



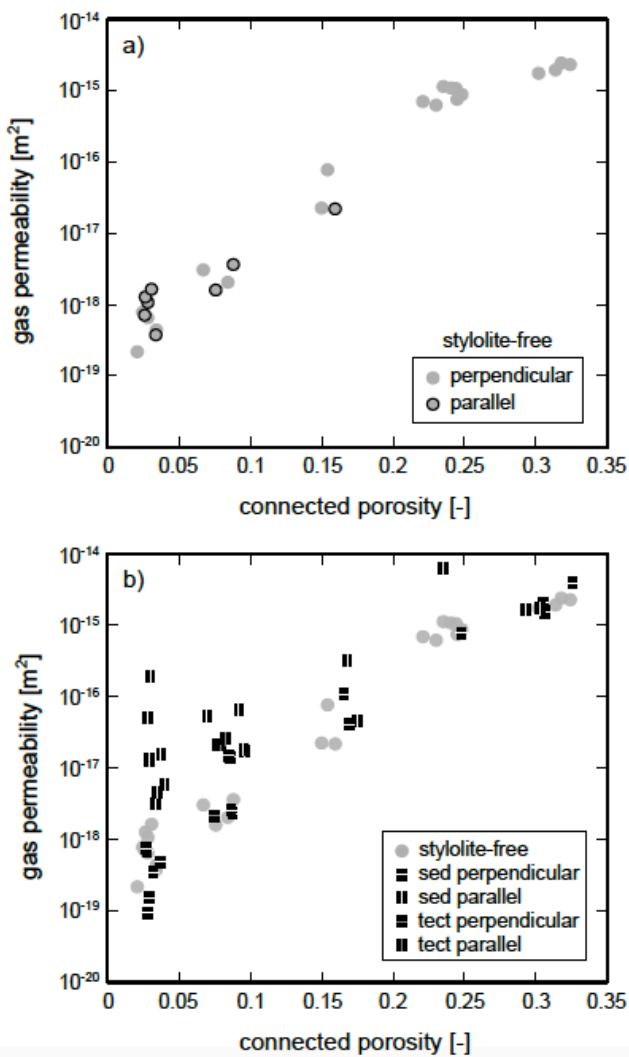
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534

535 **Figure 9.** (a) Gas permeability (measured under a confining pressure of 2 MPa) as a function of  
 536 connected porosity for intact (i.e. stylolite-free) limestone. All datapoints above a porosity of 0.2 are  
 537 taken from Lind et al. (1994). (b) Gas permeability (measured under a confining pressure of 2 MPa)  
 538 as a function of connected porosity for limestone samples containing either a stylolite parallel to  
 539 flow or a stylolite perpendicular to flow. The gas permeabilities of the intact samples (i.e. the data of  
 540 panel (a)) are also plotted in panel (b).

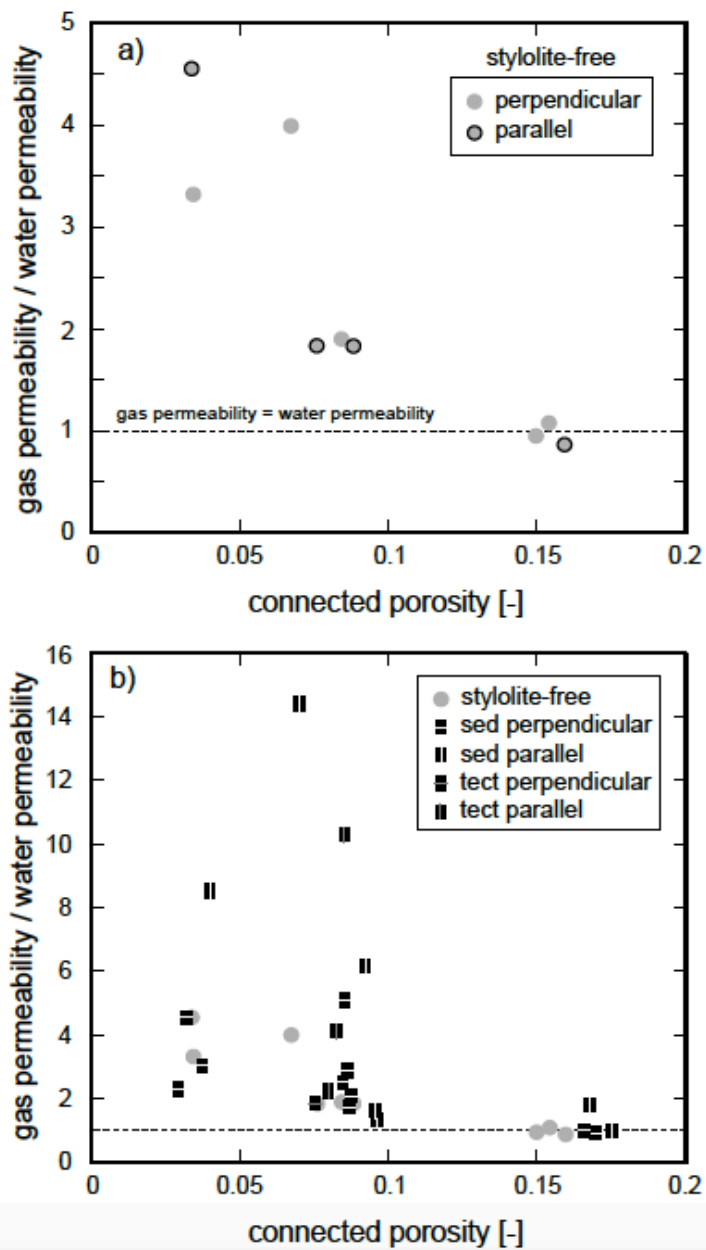
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542

543

544 **Figure 10.** (a) The ratio of gas to water permeability as a function of connected porosity for intact  
 545 (i.e. stylolite-free) limestone. (b) The ratio of gas to water permeability as a function of connected  
 546 porosity for limestone samples containing either a stylolite parallel to flow or a stylolite  
 547 perpendicular to flow. The ratios of the intact samples (i.e. the data of panel (a)) are also plotted in  
 548 panel (b).  
 549

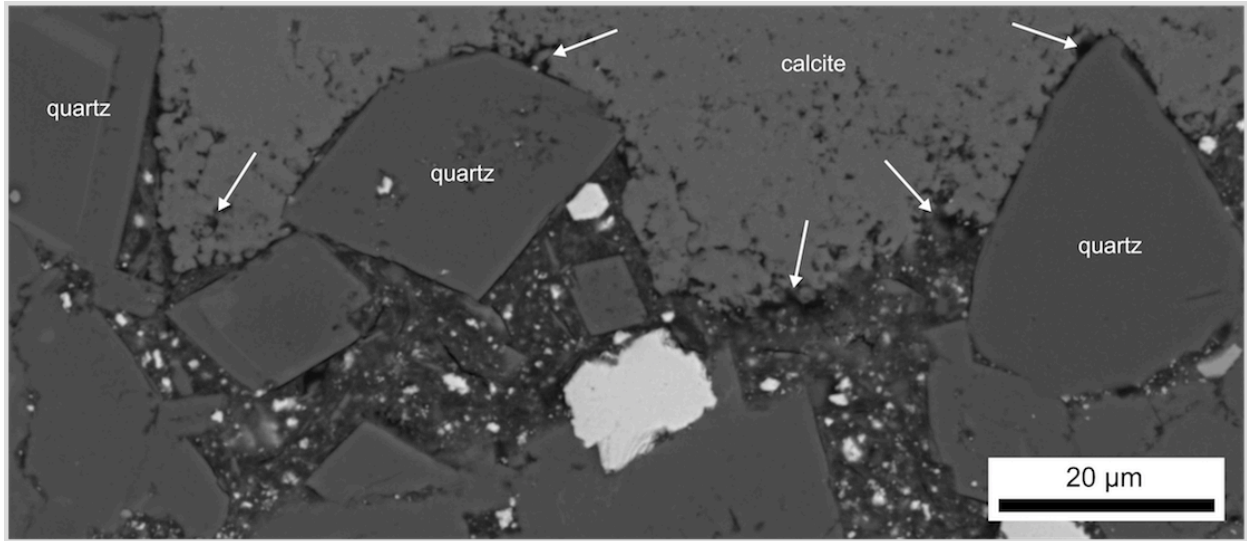


550



551 **Figure 11.** Backscattered scanning electron microscope image of the stylolite-host rock boundary  
552 (sample D3 – “Dogger” limestone from Bure). Quartz grains within the stylolite are labelled. The  
553 white arrows point to porosity (in black).

554



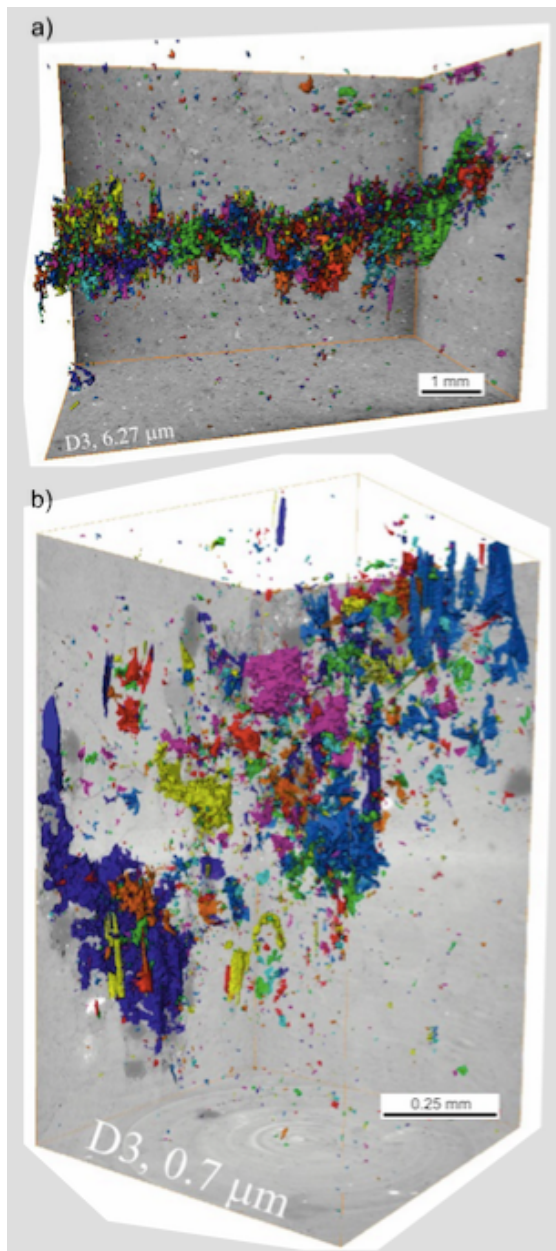
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556

557

558 **Figure 12.** Multi-resolution X-ray synchrotron microtomography images of a stylolite in sample D3  
559 (“Dogger” limestone from Bure). (a) An image of the stylolite at a voxel size of 6.27  $\mu\text{m}$ . The  
560 coloured shapes are pores; individual pores are allocated different colours. (b) An image of the  
561 stylolite at a voxel size of 0.7  $\mu\text{m}$ . The coloured shapes are pores; individual pores are allocated  
562 different colours.

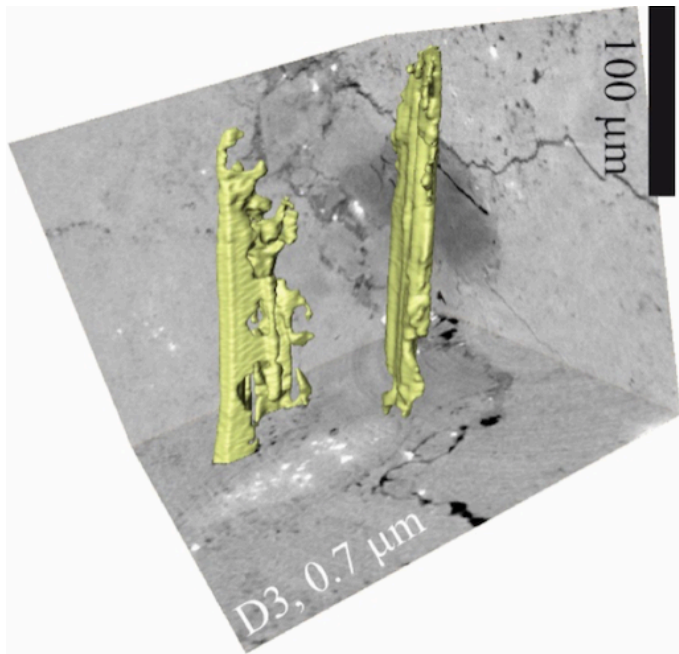
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564

565 **Figure 13.** X-ray synchrotron microtomography image showing two pores within a stylolite in  
566 sample D3 (“Dogger” limestone from Bure). The pores are “finger-like” in shape and are aligned  
567 with the teeth of the stylolite.

568



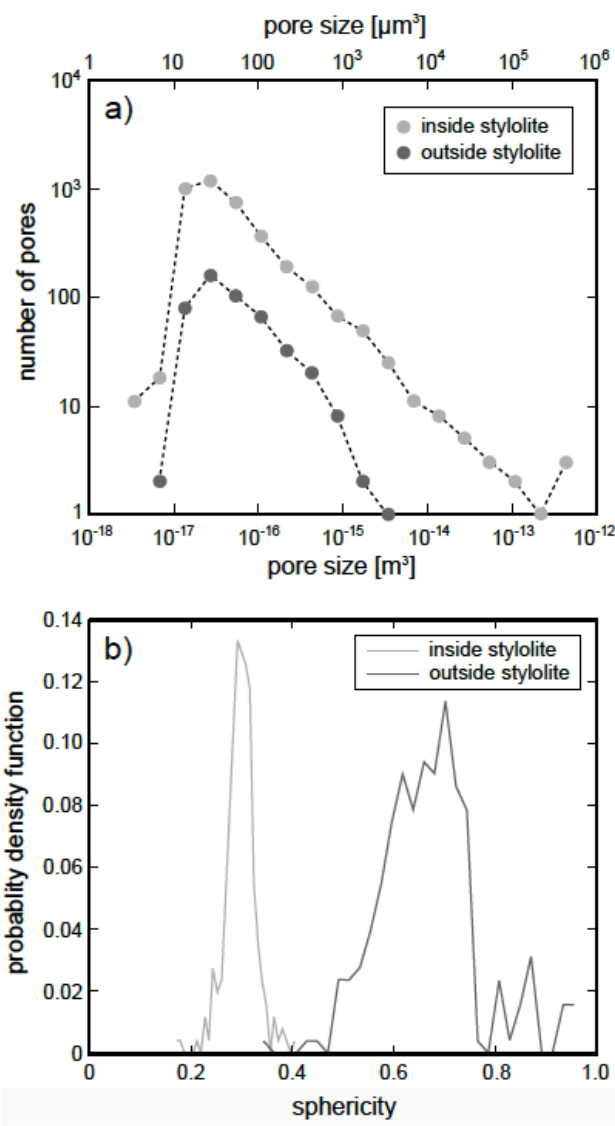
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572 **Figure 14.** X-ray synchrotron microtomography data showing (a) the number of pores within a  
 573 subvolume of 0.16 mm<sup>3</sup> as a function of pore size inside and outside of a stylolite (sample D3 –  
 574 “Dogger” limestone from Bure) and (b) the probability density function as a function of sphericity  
 575 for the pores inside and outside of a stylolite (sample D3 – “Dogger” limestone from Bure). Two  
 576 subvolumes of sample D3 with the same volume of 0.16 mm<sup>3</sup>, one inside the stylolite and one  
 577 outside of it, were used to perform these calculations.

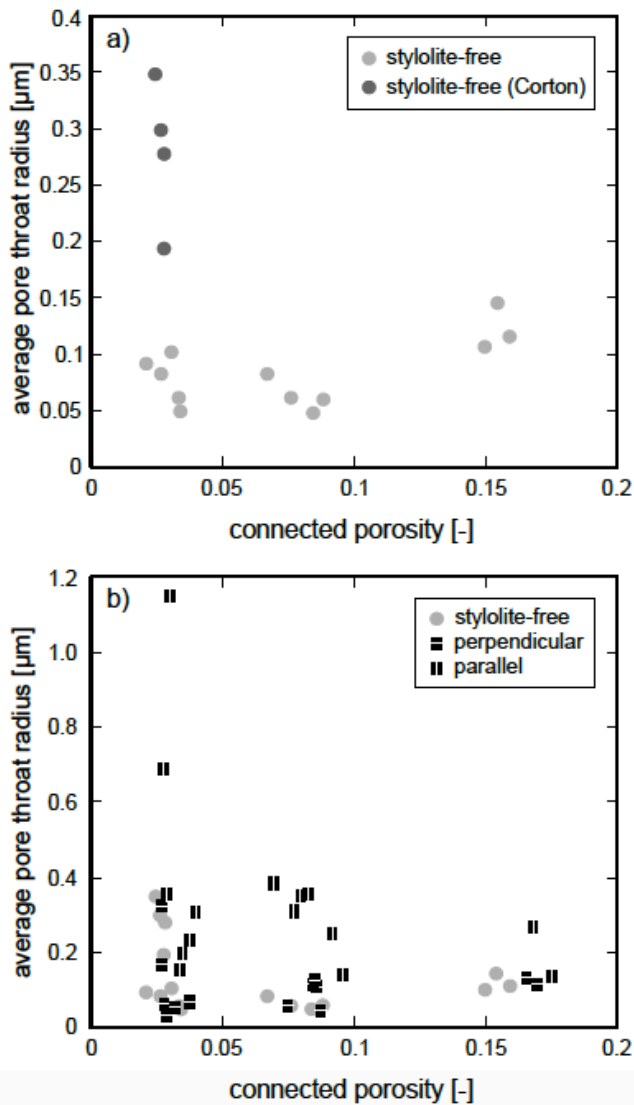
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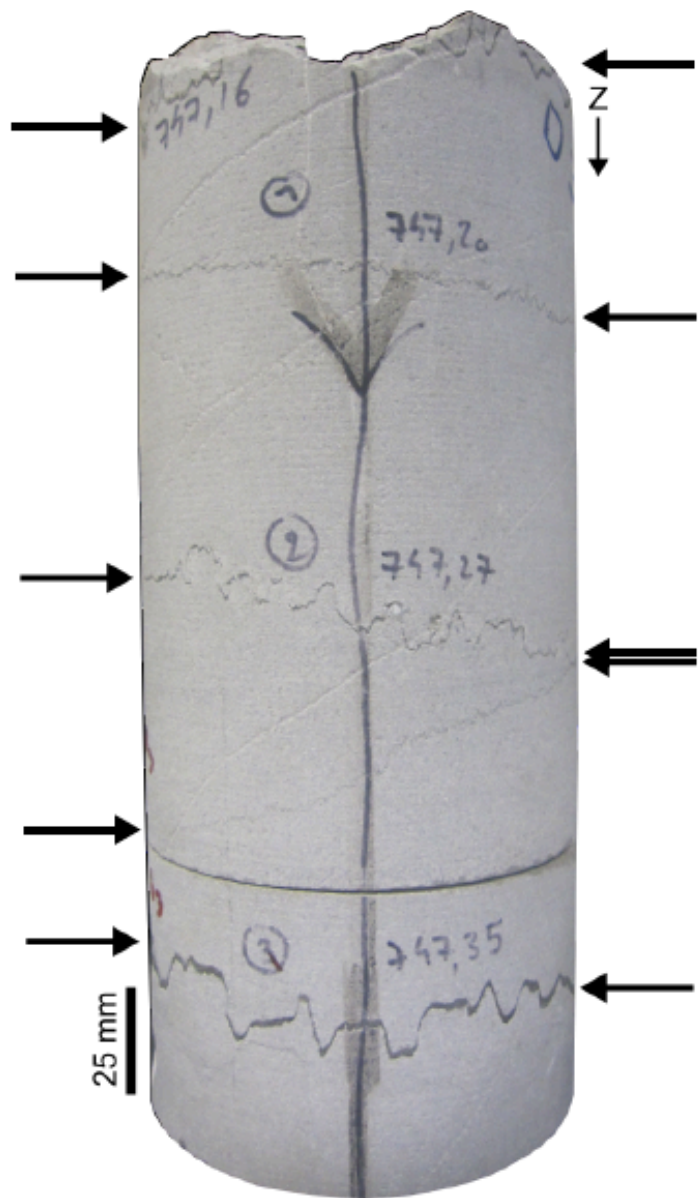
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581 **Figure 15.** (a) The average pore radius of the flow path followed by the gas molecules (calculated  
 582 using the Klinkenberg slip factor; see Equation 5) as a function of connected porosity for the intact  
 583 (i.e. stylolite-free) samples. (a) The average pore radius of the pores used by the gas particles  
 584 (calculated using the Klinkenberg slip factor; see Equation 5) as a function of connected porosity for  
 585 limestone samples containing either a stylolite parallel to flow or a stylolite perpendicular to flow.  
 586 The average pore radius of the intact samples (i.e. the data of panel (a)) are also plotted in panel  
 587 (b).  
 588



589

590 **Figure 16.** A photograph of a 78 mm-diameter core from Bure (sample D3 – “Dogger” limestone  
591 from Bure). Arrows indicate the position of sedimentary stylolites.



592

593 **Tables**

594

595 **Table 1.** X-ray powder diffraction (XRPD) analysis showing quantitative mineral composition for  
 596 the six limestones studied herein. Mineral contents of O1, O3, O6, and D3 were taken from Heap et  
 597 al. (2014).

<b>Sample</b>	<b>Stylolite-free composition (wt. %)</b>	<b>Minerals within the stylolite</b>
O1 (Oxfordian limestone)	99% calcite; <1% dolomite	dolomite, clay
O3 (Oxfordian limestone)	99% calcite, <1% dolomite; <1% gypsum; <<1% pyrite	dolomite, gypsum, pyrite, clay
O6 (Oxfordian limestone)	99% calcite, <1% dolomite; <<1% pyrite	dolomite, pyrite, clay
D3 (“Dogger” limestone)	93% quartz; 4% dolomite; 3% quartz; <<1% pyrite	dolomite, quartz, pyrite, clay
Corton limestone	99% calcite; <1% quartz	quartz, clay
Comblanchien limestone	99% calcite	clay

598

599

600 **Table 2.** Summary of the experimental data collected for this study. Porosities were measured  
601 using the triple weight water saturation technique. Gas and water permeabilities were measured  
602 under a confining pressure of 2 MPa. Gas permeabilities were measured with either argon or  
603 nitrogen gas. Water permeabilities were measured with deionised water. Asterisk indicates that the  
604 connected porosity and the gas permeability data were taken from Heap et al. (2014).

Sample	Description	Connected porosity	Gas permeability (m <sup>2</sup> )	Klinkenberg slip factor (MPa)	Water permeability (m <sup>2</sup> )	Gas/water permeability
*01	No stylo perp	0.154	$7.77 \times 10^{-17}$	0.189	$7.22 \times 10^{-17}$	1.08
*01	Sed stylo para	0.168	$3.29 \times 10^{-16}$	0.102	$1.81 \times 10^{-16}$	1.82
*01	Sed stylo perp	0.166	$1.09 \times 10^{-16}$	0.206	$1.07 \times 10^{-16}$	1.02
*03	No stylo perp	0.150	$2.25 \times 10^{-17}$	0.309	$2.37 \times 10^{-17}$	0.95
*03	No stylo para	0.159	$2.20 \times 10^{-17}$	0.286	$2.54 \times 10^{-17}$	0.87
*03	Sed stylo perp	0.169	$4.10 \times 10^{-17}$	0.278	$4.51 \times 10^{-17}$	0.91
*03	Sed stylo para	0.175	$4.65 \times 10^{-17}$	0.233	$4.78 \times 10^{-17}$	0.97
03	No stylo para	0.076	$1.60 \times 10^{-18}$	0.449	$8.72 \times 10^{-19}$	1.83
03	No stylo para	0.088	$3.63 \times 10^{-18}$	0.458	$1.98 \times 10^{-18}$	1.83
03	No stylo perp	0.084	$2.05 \times 10^{-18}$	0.568	$1.08 \times 10^{-18}$	1.90
03	Tect stylo perp	0.075	$2.14 \times 10^{-18}$	0.452	$1.16 \times 10^{-18}$	1.84
03	Tect stylo para	0.080	$2.18 \times 10^{-17}$	0.079	$9.73 \times 10^{-18}$	2.24
03	Tect stylo para	0.082	$2.62 \times 10^{-17}$	0.078	$6.35 \times 10^{-18}$	4.13
03	Tect stylo para	0.077	$2.17 \times 10^{-17}$	0.089	$7.17 \times 10^{-18}$	3.03
03	Sed stylo perp	0.087	$2.32 \times 10^{-18}$	0.621	$1.12 \times 10^{-18}$	2.07
03	Sed stylo perp	0.087	$2.61 \times 10^{-18}$	0.537	$1.51 \times 10^{-18}$	1.73
03	Sed stylo para	0.096	$1.75 \times 10^{-17}$	0.196	$1.34 \times 10^{-17}$	1.31
03	Sed stylo para	0.095	$1.84 \times 10^{-17}$	0.195	$1.15 \times 10^{-17}$	1.60
*06	No stylo perp	0.067	$3.04 \times 10^{-18}$	0.335	$7.62 \times 10^{-19}$	3.99
*06	Sed stylo para	0.070	$5.36 \times 10^{-17}$	0.102	$3.72 \times 10^{-18}$	14.41
*06	Sed stylo para	0.092	$6.58 \times 10^{-17}$	0.111	$1.07 \times 10^{-17}$	6.15
*06	Sed stylo	0.084	$1.51 \times 10^{-17}$	0.241	$6.04 \times 10^{-18}$	2.50



	perp					
*O6	Sed stylo perp	0.086	$1.43 \times 10^{-17}$	0.252	$4.98 \times 10^{-18}$	2.87
*O6	Sed stylo perp	0.085	$1.39 \times 10^{-17}$	0.217	$2.73 \times 10^{-18}$	5.09
*D3	No stylo perp	0.034	$4.38 \times 10^{-19}$	0.557	$1.32 \times 10^{-19}$	3.32
*D3	No stylo para	0.034	$3.69 \times 10^{-19}$	0.447	$8.11 \times 10^{-20}$	4.55
*D3	Sed stylo perp	0.037	$4.88 \times 10^{-19}$	0.398	$1.61 \times 10^{-19}$	3.03
*D3	Sed stylo perp	0.032	$3.44 \times 10^{-19}$	0.521	$7.55 \times 10^{-20}$	4.56
*D3	Sed stylo perp	0.029	$1.56 \times 10^{-19}$	0.847	$6.83 \times 10^{-20}$	2.28
*D3	Sed stylo para	0.040	$5.98 \times 10^{-18}$	0.089	$7.01 \times 10^{-19}$	8.53
COMB	No stylo para	0.031	$1.64 \times 10^{-18}$	0.270	-	-
COMB	No stylo para	0.026	$7.20 \times 10^{-19}$	0.335	-	-
COMB	Sed stylo para	0.037	$1.54 \times 10^{-17}$	0.118	-	-
COMB	Sed stylo para	0.034	$3.20 \times 10^{-18}$	0.180	-	-
COMB	Sed stylo para	0.035	$4.64 \times 10^{-18}$	0.140	-	-
COMB	No stylo perp	0.021	$2.18 \times 10^{-19}$	0.298	-	-
COMB	Sed stylo perp	0.027	$6.95 \times 10^{-19}$	0.164	-	-
CORT	No stylo para	0.026	$1.28 \times 10^{-18}$	0.092	-	-
CORT	No stylo para	0.028	$1.07 \times 10^{-18}$	0.099	-	-
CORT	Sed stylo para	0.033	$1.98 \times 10^{-16}$	0.024	-	-
CORT	Sed stylo para	0.030	$5.22 \times 10^{-17}$	0.040	-	-
CORT	Sed stylo para	0.031	$1.31 \times 10^{-17}$	0.078	-	-
CORT	No stylo perp	0.026	$7.85 \times 10^{-19}$	0.079	-	-
CORT	No stylo perp	0.029	$6.58 \times 10^{-19}$	0.142	-	-
CORT	Sed stylo perp	0.027	$7.48 \times 10^{-19}$	0.085	-	-
CORT	Sed stylo perp	0.031	$9.45 \times 10^{-20}$	0.433	-	-

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