

1 **Variability in spatial distribution of mineral phases in the Lower Bowland**
2 **Shale, UK, from the mm- to μm -scale: quantitative characterization and**
3 **modelling**

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31 **Abstract**

32 The microstructure of a highly laminated Lower Bowland Shale sample is characterized at
33 the micron- to millimeter scale, to investigate how such characterization can be utilized for
34 microstructure-based modelling of the shale's geomechanical behavior. A mosaic of scanning
35 electron microscope (SEM) back-scattered electron (BSE) images was studied. Mineral and
36 organic content and their anisotropy vary between laminae, with a high variability in
37 fracturing and multi-micrometer aggregates of feldspars, carbonates, quartz and organics. The
38 different microstructural interface types and heterogeneities were located and quantified,
39 demonstrating the microstructural complexity of the Bowland Shale, and defining possible
40 pathways for fracture propagation. A combination of counting-box, dispersion, covariance
41 and 2D mapping approaches were used to determine that the total surface of each lamina is 3
42 to 11 times larger than the scale of heterogeneities relative to mineral proportion and size.
43 The dispersion approach seems to be the preferential technique for determining the
44 representative elementary area (REA) of phase area fraction for these highly heterogeneous
45 large samples, supported by 2D quantitative mapping of the same parameter. Representative
46 microstructural models were developed using Voronoï tessellation using these characteristic
47 scales. These models encapsulate the microstructural features required to simulate fluid flow
48 through these porous Bowland Shales at the mesoscale.

49

50 **Highlights**

- 51 • The microstructure of the Lower Bowland Shale was studied quantitatively.
- 52 • The presence of multi-scale microstructural interfaces supports hydraulic fracturing
53 potential in the Bowland Shale.
- 54 • The dispersion approach is the most suitable method to define representative
55 elementary areas.
- 56 • 2D mapping of phase area fraction supports the dispersion approach.
- 57 • A representative model of the geometry of the microstructure based on Voronoi
58 tessellation was generated.

1 **1. Introduction**

2 Significant shale gas and oil resources are believed to exist in Western Europe (EIA, 2013)
3 and in particular in the UK (EIA, 2015a), such as in Carboniferous and Jurassic-Age shale
4 formations. One example is the Bowland Basin, in the west portion of the Pennine Basin,
5 close to Blackpool, Lancashire, Northern England (EIA, 2015). The Bowland Shale is
6 generally divided into two units: the Upper Bowland Shale composed of thick layers of
7 marine-deposited organic matter-rich mudstone, whilst the Lower Bowland Shale is
8 composed of thinner alternating layers of organic-matter-poor and organic-matter rich
9 mudstone (Andrews, 2013).

10 The Upper Bowland Shale demonstrates similarities to North American shale gas plays,
11 whereas relatively few regions in the Lower part have been identified as potentially
12 productive. The Lower Bowland Shale is still largely undrilled and its geographical extent is
13 currently uncertain (Andrews, 2013). Despite this lack of data, the potential of the Lower
14 Bowland Shale is considered to be important, albeit with a higher uncertainty than the Upper
15 Bowland Shale (Andrews, 2013). Shale gas potential depends on the oil and gas retention
16 capacity of the system, which is directly related to the microstructure: the nature and the
17 specific adsorption of organic and inorganic phases, their structural relationships, and those
18 of the pore and fracture networks. Moreover, quantifying the relationships between porosity,
19 permeability, pore size distribution and brittleness in combination with the mineralogy and
20 rock fabric is essential for the estimation of shale gas potential (Josh et al., 2012).
21 Microstructural heterogeneities such as variations of grain size, shape, mineralogy, elasticity,
22 anisotropy and stiffness, together with preexisting defects can create local stress
23 concentrations. Such stress concentrations influence the initiation and behavior of fractures in
24 shales, such as hydraulic fractures (Keneti and Wong 2010), mechanical fractures (Van de

25 Steen et al., 2003) and desiccation fractures (Hedan et al., 2012; Fauchille et al., 2016;
26 Figueroa Pilz et al., 2017) in response to the local stress field. In a general sense, Sone and
27 Zoback (2013) and Amann et al., (2014) have shown how significantly the microstructure can
28 impact upon the mechanical properties of shale. As a direct result, the microstructure
29 therefore influences the gas retention process (Bernard et al., 2010). Despite the strong
30 interest in the Lower Bowland Shale in the UK, its petrophysical properties and
31 heterogeneities are still poorly understood, but are expected strongly to influence yield during
32 any gas production.

33 Due to the sub-micrometer scale of shale components, scanning electron microscopy (SEM)
34 and transmission electron microscopy (TEM) are often used to characterize the
35 microstructural features of shales (Ma et al., 2017). Houben et al., (2014, 2016) and Klaver et
36 al., (2015) constructed mosaics of high resolution SEM images to quantify the morphology
37 and size distribution of pores, clay and organic matter particles in the Opalinus Clay
38 (Switzerland) and Posidonia Shale (Germany), and the microstructure of European Early
39 Jurassic Shales. However, in light of the well-known heterogeneity of such rocks and the
40 dependence of large-scale features (clay minerals, organics) on the structure of fine-scale
41 features (clusters of inorganic grains, clay phase, fractures), the size of the field of view is
42 often limited by the compromise between sample size and spatial resolution. Fracture
43 initiation may depend on heterogeneities down to the nanometer scale if differential stresses
44 are high enough, but fracture propagation is affected by multi-scale heterogeneities (Griffith,
45 1924; Jaeger and Cook, 1976). Consequently, the characterization of “low-scale” (meso- and
46 macro-scales) heterogeneities should be considered in fracture propagation studies.

47 Mechanical behavior of shale at the macroscale is strongly dependent on the behavior at
48 smaller scales, for example due to the primary structure and arrangement of particles
49 (anisotropy), and also to the variability of small scale components such as organic matter,

50 rigid inclusions and clay minerals (Sayers, 1993; Wang, 2012; Wang et al. 2015, Bonnelye et
51 al., 2017 a, b). As a consequence, understanding the microscale structure may aid detailed
52 comprehension of the macroscale behavior and hence bulk constitutive relations. To predict
53 strain location and fracturing behavior at the macroscale, simple models of grain-based
54 microstructure taking into account microstructural parameters such as grain size, shape,
55 proportion, elongation ratio and orientation ratio are required (van den Eijnden et al., 2015,
56 2016, 2017).

57 This study we investigates the variability of microstructural parameters (phase area fraction,
58 grain size, shape, elongation and orientation) from the mm to μm scales in a sample of the
59 Lower Bowland Shale, with a millimeter mosaic of SEM images at sub-micrometer
60 resolution where two laminae were identified. The goals of the study are:

- 61 i) to quantitatively characterize the spatial variability of mineral phases of the Lower
62 Bowland shale;
- 63 ii) to describe the heterogeneity of the microstructure using several methods allowing
64 quantifications of representative elementary areas;
- 65 iii) to establish a simple representative model of the Lower Bowland shale
66 microstructure based on the approach of van den Eijnden et al. (2015-2017),
67 which takes into account (i) and (ii).

68 **2. Geological Setting and Sampling**

69 The Carboniferous Bowland Shale occurs across the center and north of England. Its
70 occurrence extends from Merseyside to Humberside and Loughborough to Pickering
71 (Andrews, 2013). It is divided into an upper unit with thick and continuous (a few hundreds
72 of meter thick) shale formations, and a lower unit containing shale interbedded with clastic
73 and carbonate deposits, explained by the influence of glacio-eustatic sea levels changes and

74 tectonic events (Gawthorpe, 1987; Andrews, 2013). For this study, one sample of Lower
75 Bowland Shale was collected from the Preese Hall-1 borehole at a depth of 2495.27 meters,
76 supplied by the British Geological Survey (BGS) in Nottingham (UK). The Preese Hall-1
77 borehole was drilled in 2010 on the Fylde coast of NW Lancashire at 53° 49' 19.006"N; 2°
78 56'56.576" near to Blackpool, north-west England. Preese Hall-1 was the first dedicated
79 unconventional shale gas borehole drilled in the UK and Europe (de Pater and Baisch, 2011).
80 The sample chosen for this study corresponds to sample B8 in Fauchille et al. (2017). The
81 sample has a laminated microtexture and the mean organic content (TOC) is 1.1 wt %,
82 measured with a Leco carbon analyzer using 100 mg powder samples, at the University of
83 Newcastle (UK). Centimeter-size samples were used for making the powder samples, to
84 ensure several laminae are included in the powder. According to bulk XRD measurements
85 (the accuracy of quantification of XRD results in terms of modal proportions is around 1%),
86 the sample is composed of 51.7 wt% quartz, 18.4 wt% kaolinite, 11.1 wt% ankerite, 8.6 wt%
87 albite, 5.1 wt% muscovite, 3.5 wt% calcite and 1.6 wt% pyrite. The intact sample was
88 vacuum impregnated with low-viscosity epoxy resin (Araldite 2020) at room temperature
89 (20°C) and mechanically polished as a thin section with carbide and diamond products for
90 scanning electron microscopy.

91 **3. Methods Employed**

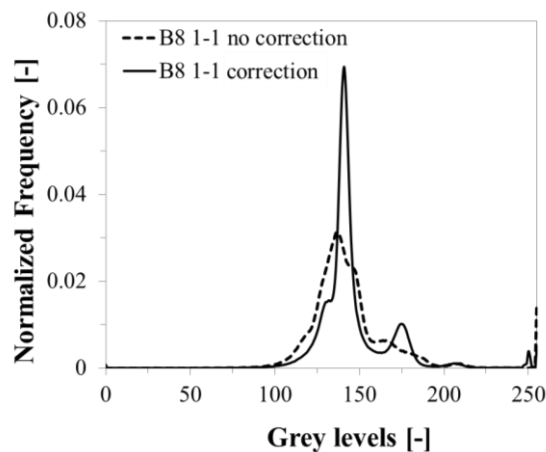
92 **3.1. Scanning electron microscope (SEM) mosaic image acquisition**

93 A mosaic of 4×11 back-scattered electron (BSE) images each of 1280 × 960 pixels was
94 acquired from the carbon-coated polished specimen at a resolution of 0.4 μm.pixel⁻¹ (Jeol
95 JSM-6610LV) with 23% overlap (Klaver et al., 2012; Vergès and Morales, 2014; Fauchille,
96 2015). The mosaic is 4204 × 8128 pixels (8bit) covering 1.68 × 3.25 mm². The working
97 distance (WD) was 10 mm and the beam accelerating voltage was 20 kV. A magnification of

98 250X proved to be a good compromise between spatial resolution, field of view and
99 acquisition time, and minimized drift of the incident electron beam and scanning lens.
100 Contrast and brightness were adjusted to have the organic matter particles in black (0) and the
101 heavy minerals in white (255). The parameters were optimized to obtain a histogram with
102 distinct peaks to facilitate mineral segmentation (Prêt et al., 2010a; Robinet et al., 2012,
103 Fauchille, 2015).

104 3.2. Correction and segmentation of images

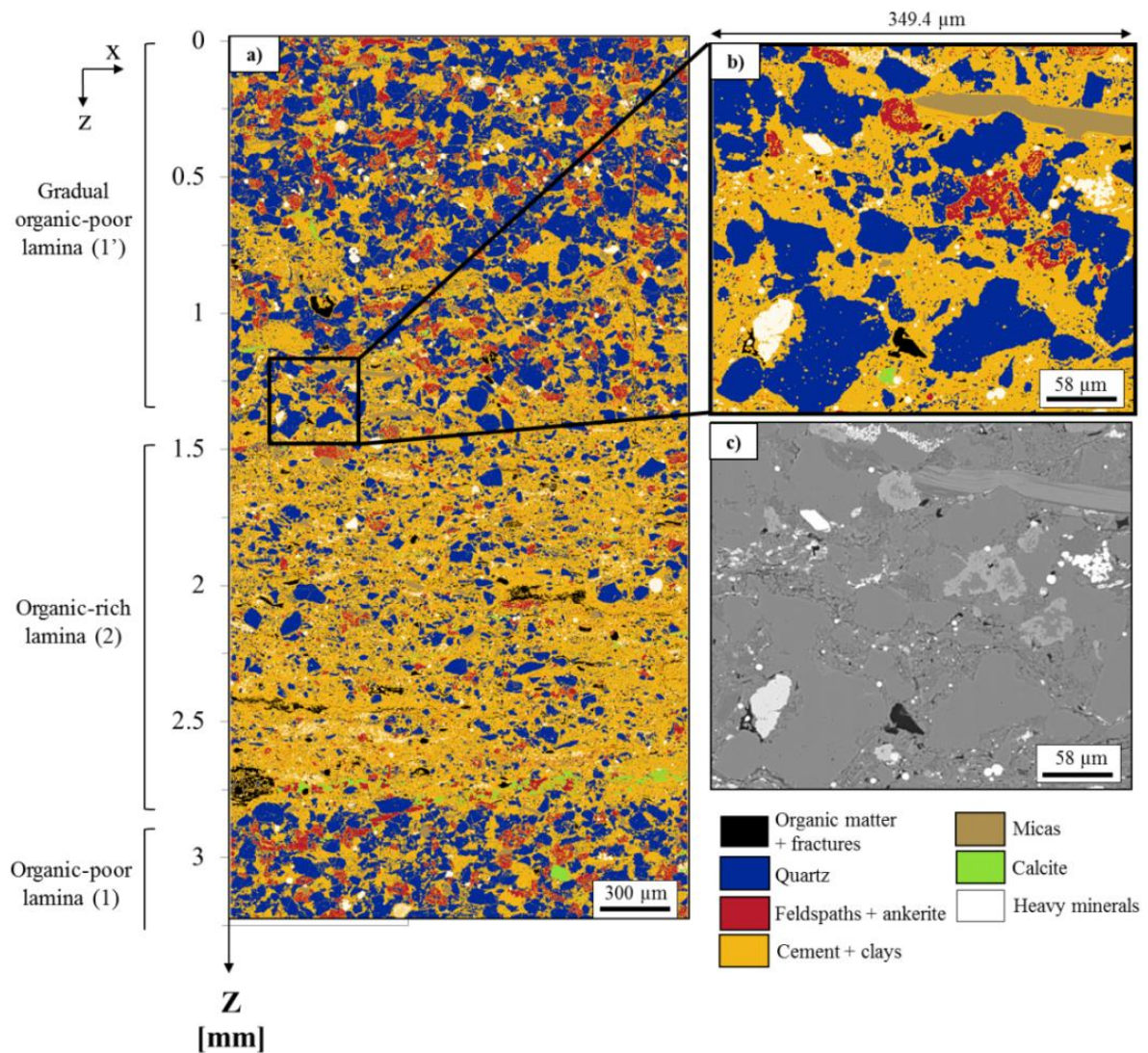
105 All images were drift-corrected to produce a homogeneous histogram across all images of the
106 mosaic. A linear gray level drift was observed in the x direction (horizontal) on each image,
107 due to slight shadowing of the incident beam caused by the low-vacuum aperture of the pole-
108 piece. Images were corrected in vertical sections of 30×960 pixels to balance the gray
109 levels, producing consistent histograms with well-separated peaks (see Fig. 1). No significant
110 shift was detected in the z direction (vertical).



111
112 **Fig. 1. Gray level histogram of the first image of mosaic B8 1-1 (this image corresponds**
113 **to the first column and first line of the mosaic) before and after drift correction.**

114 The mineral phases were segmented on the BSE images using a combination of filters and
115 gray level thresholding using in-house codes written for Visual Studio, Matlab and ImageJ

116 (Peters, 2009; Prêt et al., 2010a,b; Robinet et al., 2012; Khan et al., 2014). The organic matter
117 and fractures (om+fractures), feldspars (felds), ankerite, calcite (carb) and heavy minerals (h-
118 min) were only segmented by gray levels with ImageJ. The gray levels used for thresholding
119 were: 0 to 64 for organic matter and fractures, 65 to 133 for clays and quartz cement, 134 to
120 147 for quartz inclusions, 148 to 163 for micas, 164 to 192 for feldspars and ankerite, 193 to
121 218 for calcite and 219 to 255 for heavy minerals. Due to their similar silica contents, the
122 segmentation of quartz and micas was performed using the gray level histogram and a local
123 variance filter (Prêt et al., 2010a, Robinet et al., 2012). In ImageJ software, the value of the
124 variance was chosen at 10 to detect the homogenous areas of quartz, according to the BSE
125 images. Owing to the small size of kaolinite particles and their silica content being close to
126 the background, clays were removed in the same phase as the quartz cement to form the
127 “binder” of the sample. To improve the segmentation of individual grains, the 1-pixel
128 boundaries of quartz and feldspars grains, were moved to the background. This implies that
129 the segmentation of grains may be underestimated but better separated for calculating
130 morphological parameters. The segmentation, shown in Fig. 2 takes into account the
131 separation between the large grains of quartz and the cement, but cannot differentiate
132 between organic matter particles and fractures, both of which present the same gray scale
133 value (black) on BSE images. The fractures segmented on the mosaic are filled by organic
134 matter or empty. Fractures can be generated by pore fluid overpressuring through organic
135 decomposition and petroleum generation, by the late migration of organic matter. Open
136 cracks can be produced as a result of sample depressurization during recovery and during the
137 sample preparation (Fauchille et al., 2017). There is a fracture filled by ankerite (top right
138 part in Figure 2), which was therefore segmented as ankerite minerals (carbonates).



139

140 **Fig. 2.** (a) Mineral phase segmentation on the whole mosaic of SEM-BSE images of the
 141 **Bowland Shale** sample. (b) Magnified view of a region of (a). (c) **Original**
 142 **(unsegmented) back-scattered electron image** corresponding to (b).

143 **3.4. Image analysis**

144 **Microstructural parameters**

145 The anisotropy of the microstructure was studied using the 2D microstructural parameters
 146 used on shale (Robinet et al., 2012; Fauchille et al., 2014, 2016). These are the area fraction
 147 (ratio between the number of pixels of a grain type and the total number of pixels in the

148 calculation window), the grain size (number of pixels in one grain), the elongation ratio and
149 the orientation, calculated with the ellipsoid plugin in ImageJ software. The ellipsoid axial
150 ratio and orientation of each grain with respect to the reference horizontal axis was
151 calculated, but only for grains larger than 10 pixels, in order to avoid any resolution artefacts.

152 **Determination of correlation lengths of the microstructure**

153 The estimation of a representative elementary area (REA) (also called homogenization
154 surface) of a material for a specific property, scale and resolution (Bear, 1972; Bear and
155 Bachmat, 1984; Grolier et al., 1991; Al-Raoush et al., 2010), can be problematic in shales due
156 to their well-known multi-scale and multi-modal heterogeneities. However, the
157 characterization of heterogeneities and the identification of REAs are crucial for upscaling
158 and modelling studies. In theory, a surface is representative of a parameter at a specified scale
159 when this parameter becomes independent of the size of the field of view, including
160 statistically all types of structural heterogeneities (Bear, 1972; Kanit et al., 2003; Al-Raoush
161 et al., 2010). In this paper, three methods were used and compared to estimate representative
162 elementary surfaces of the microstructure and to identify its heterogeneities: (i) the counting-
163 box method, (ii) the dispersion approach, and (iii) the covariance. All calculations were
164 performed with in-house codes in Visual-Studio (C) and Matlab. Other methods and
165 parameters were used to extract correlation lengths from an image, such as the entropy
166 method (Boger et al., 1992) and the percolation method (Cosenza et al., 2015), but they are
167 not taken into account in this paper.

168 ***Counting box method***

169 A simple way to approximate the REA is the “counting-box method”, that considers a
170 succession of increasing-size domains until the parameter measured is constant
171 (VandenByggaert and Protz, 1999; Kameda et al., 2006; Houben et al., 2014), plus or minus a

172 defined confidence limit which was chosen to be 10% in our case, consistent with previous
173 studies on geological materials (VandenByggaert and Protz, 1999; Houben et al., 2014; Vik
174 et al., 2014). In this study, each lamina was defined over an area of 4404×3302 pixels (1.76
175 $\times 1.32$ mm), and the domains between 10×10 pixels ($4 \times 4 \mu\text{m}$) to 3302×3302 pixels
176 (1.32×1.32 mm) were used for this method. The first domain was chosen in the center of
177 each lamina, in order to have the maximum number of possible domains.

178 *Dispersion approach*

179 The dispersion approach was introduced by Kanit et al., (2003). Applied to a 2D image, it
180 involves dividing the image into equal subsamples, in which a chosen parameter is
181 calculated. The goal of this approach is to determine the number of subsamples needed limit
182 to an acceptable level the relative error of this parameter for the set of subsamples.

183 The mosaic of BSE images was divided into independent square domains D from 10×10 to
184 1876×1876 pixels (4×4 and $750 \times 750 \mu\text{m}^2$, respectively) (Kanit et al., 2003; Keller et al.,
185 2013). Kanit et al., (2003) used this approach to divide random and Voronoï mosaics into a
186 finite number of cells. In this paper, the domains D are the finite square cells of the
187 experimental mosaic. This mosaic cannot be extended therefore the number of D decreases as
188 their size increases. Each domain is thus a subsample of the mosaic and contains a finite
189 number of grains. The pixels of the domains are independent from one domain to another, but
190 it is possible that long or large grains (such as micas, Fig. 2b) can be included in multiple
191 neighboring domains. The standard deviation, σ_D , of the phase area fraction $P_D(x, z)$ on each
192 lamina as a function of domain size D was used to quantify the heterogeneity of the
193 microstructure (Equations 1 and 2) with a statistical approach.

$$P_D(x, z) = \frac{1}{D} \sum_{n=x-\frac{D-1}{2}}^{n=x+\frac{D-1}{2}} \sum_{m=z-\frac{D-1}{2}}^{m=z+\frac{D-1}{2}} G_{(n,m)} \quad (1)$$

194 $P_D(x, z)$ is the area fraction of a defined phase (examples: carbonates, quartz,
 195 cement/clays...) in a domain, D , centered at (x, z) . $P(x, z)$ varies between 0 and 1. D is the
 196 size of the domain in pixels (and is always an odd number). G is a binary function (0 or 1). G
 197 = 1 when the pixel (n, m) corresponds to the chosen phase, and 0 when the pixel corresponds
 198 to a different phase. For example, when the area fraction of calcite is calculated: $G_{(n,m)} = 1$ if
 199 the pixel (n, m) corresponds to calcite, and 0 if the mineral is not calcite.

200 To have a whole view of the dispersion of area fraction values, $P_D(x, z)$ was mapped in 2D
 201 increments by 1 pixel in the x and z directions over the mosaic. The standard deviation σ_P of
 202 $P_D(x, z)$ was then calculated as a function of domain size (Equation 2):

$$\sigma_P = \sqrt{\frac{\sum (P_D(x, z) - \bar{P}_D)^2}{N_D}} \quad (2)$$

203 \bar{P}_D is the mean value of the phase area fraction in all domains D and N_D is the number of
 204 domains D .

205 This dispersion approach allows us: (i) to identify the minimum size of a representative area
 206 of area fraction with $\sigma_P < 0.1$; (ii) to map in 2D the area fraction to present a direct
 207 visualization of microstructural variability and its multi-scale heterogeneities; and (iii) to
 208 investigate the evolution of the variance σ_P^2 as a function of D in order to estimate the effect
 209 of the randomness of the microstructure on the whole mosaic. The characterization of

210 heterogeneities and REAs will be used for microstructural modelling afterwards. Other
211 parameters of grains, such as the size, the representative ellipse axial ratio or its long axis
212 orientation can be investigated to map the spatial variability of microstructure, but the area
213 fraction was chosen here as a good indicator of the microstructure, and it is commonly used
214 in 2D and 3D texture analysis (VandenByggart et al., 1999; Madi et al., 2005; Houben et al.,
215 2014; Fauchille, 2015; Klaver et al., 2015).

216 ***Covariance***

217 The covariance approach is commonly used to determine characteristic scales and to quantify
218 the anisotropy of 2D microstructure (Berryman and Blair, 1986; Stoyan et al., 1995; Jeulin,
219 2001; Kanit et al., 2003; Degallaix and Ilschner, 2007; Rolland du Roscoat et al., 2007;
220 Rudge et al., 2008; Gaboreau et al., 2016). The principle of covariance is the measurement of
221 how well one image matches a spatially shifted copy of itself. The covariogram
222 $C(X, h)$ measures the intersection of a 2D image X and the translated image of X by a
223 distance h (Kanit et al., 2003) (Equation 3):

$$C(X, h) = \int c(x)c(x + h) dx \quad (3)$$

224 where $c(x)$ is the phase detection function, defined by :

$$c(x) = \begin{cases} 1 & \text{if } x \in X \\ 0 & \text{else} \end{cases}$$

225 The dispersion of the state of two points x_1 and x_2 between the image X and the image X
226 translated by h can therefore be quantified (Kanit et al., 2003). For a non-periodic
227 microstructure, correlations between points decrease with increasing h . When h is
228 sufficiently large, there is no correlation anymore between the points of the two images.
229 Consequently, $C(X, h)$ describes an asymptotic behaviour limited at the mean phase

230 proportion, \bar{P}^2 , of the whole image. When the asymptote is not reached, the points included in
231 the image are partially correlated, and therefore the image size is not considered to be
232 representative of its content. The distance h corresponding to the scale beyond which the
233 asymptote matches will be therefore considered as an indicator for defining an adequate scale
234 of microstructure. The covariograms of the parts corresponding to each lamina (independent
235 areas of 4404×3302 pixels) were calculated. All phases were first considered independently
236 in the calculation.

237 **3.5. Numerical model for microstructure and mineral geometry**

238 A simple model for convex grains, based on Voronoï diagrams (Sellers and Napier, 1997;
239 Fan et al., 2004; van den Eijnden et al., 2015, 2017) is used to reproduce the microstructure.
240 Previously this model has been used to generate simple microstructures in a periodic
241 representative elementary volume with straightforward finite element discretization (van den
242 Eijnden, 2015), and to model the microstructure of a Callovo-Oxfordian claystone (van den
243 Eijnden et al. 2017). Here, the model is used to investigate microstructures with geometrical
244 properties matching those of the different laminae of the studied Bowland shale. The model
245 applies Voronoï tessellation on a set of randomly generated sites and uses the resulting
246 diagram as a geometrical model of the microstructure. Preferred orientations and grain
247 elongation are introduced by stretching the domain during tessellation by an amount ξ which
248 thereby becomes a model parameter for the grain shape contribution to total anisotropy. The
249 model calibration is by a minimization of the root-mean-square error between the model
250 distributions and experimental distributions of total grain elongation L and orientation β (see
251 Fig. 6). As the model is based on Voronoï tessellation, there is little flexibility in the grain
252 size distribution without a strong distortion of the grain geometry distributions. For this
253 reason, no attempt was made to reproduce the grain size distributions, and the emphasis here
254 is on reproducing the correct distributions of (1) elongation (also called length ratio), (2)

255 grain orientation and (3) area fraction of all mineral groups (including organic matter and
256 fractures). Calibration of the model has indicated that grain circularity corrections, as part of
257 the model in van den Eijnden et al., (2017), are not needed to capture the grain geometry
258 distributions of the shale.

259 Distributions of grain orientation and elongation for the different mineral types can be
260 reproduced reasonably well using simple Voronoï tessellation while slight manipulation of
261 the sites and stretching of the domain is applied during the tessellation operation. Based on
262 the parameter distributions and area fractions of the different mineral types, mineral
263 properties can be assigned. Their parameter distributions are then approached in the model.

264 The distributions of orientation and elongation are matched in an average distribution of all
265 phases combined (quartz, carbonates, feldspars, micas, heavy minerals, organics and
266 fractures). In the microstructure model, the phases are scattered as a function of their
267 experimental area fraction. The cement and clays were first divided into random cells but
268 their boundaries were then merged to form a unique phase. van den Eijnden et al., (2017)
269 provide further details on the method of developing the microstructure model.

270 According to the experimentally determined area fraction of mineral phases, the mineral
271 types were assigned following the conditional probability $P_M(m_i|L, \beta)$ to meet specified
272 orientation and elongation properties, matching the distributions in Fig. 11. m_i is the type of
273 mineral phase, L the elongation and β the orientation, which is the direct application of
274 Bayes' theorem to derive a conditional probability, with an additional correction factor to
275 account for grain-size dependency of the model (Bernardo and Smith, 2001; Berger, 2013).
276 This probability is defined by Equation 4:

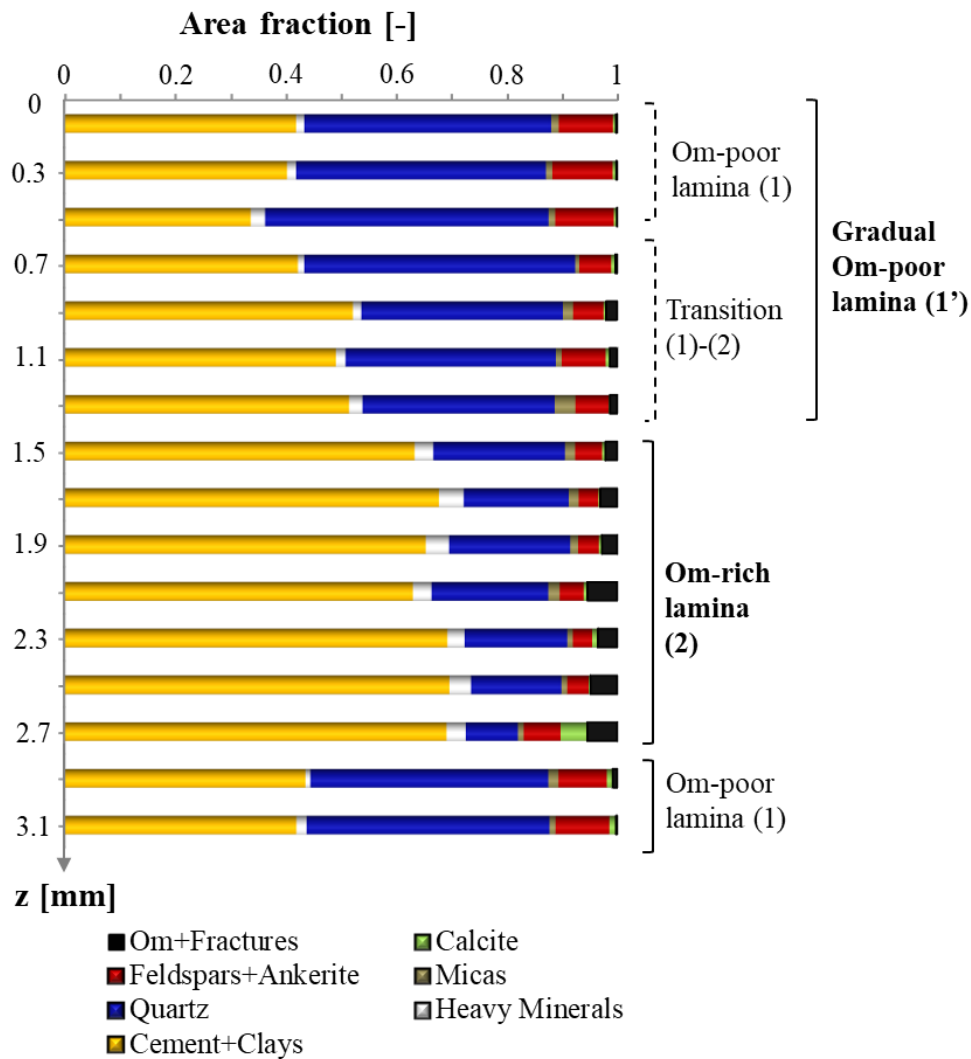
$$P_M^N(m_i|L, \beta) = \frac{q_D^N(L, \beta|m_i)f_D^S(m_i)}{q_M^N(L, \beta)} c_i \quad (4)$$

277 with $f_D^S(m_i)$ being the area fraction of mineral phase m_i (following Table 1), $q_D^N(L, \beta|m_i)$ is
 278 the bivariate distribution of L and β in the data for phase type m_i and $q_M^N(L, \beta)$ is the
 279 bivariate distribution of L and β of all phases in the model. c_i is a ratio to correct for the
 280 influence of the correlation between grain size and grain geometry in the model on the final
 281 area fractions of the minerals. The values of c_i are defined theoretically as the ratio of the
 282 resulting model area fraction over the expected area fraction of the assigned materials, and
 283 requires the distribution of model grain size as a function of L and β . However, here the
 284 ratios c_i have been determined based on the resulting area fractions of the model and updated
 285 iteratively to obtain the required area fractions. Values for c_i range between 0.6 and 1.25.

286 **4. Results**

287 **4.1. Vertical variability of phase content**

288 Fig. 3 shows the vertical variation of all area fractions, for independent horizontal domains of
 289 $200 \mu\text{m} \times 1.682 \text{ mm}$. A width of $200 \mu\text{m}$ was chosen as a good compromise to show
 290 quantitatively the vertical variability of area fractions including different types of
 291 microstructural heterogeneities.



292

293 **Fig. 3. Vertical variability of the area fraction of each mineral along the z axis of the**
 294 **mosaic, based on horizontal domains of 500 × 4204 pixels (200 μm × 1.682 mm).**

295 Three different regions were identified:

296 **(1) Organic-poor laminae** ($z = 0-0.80$ mm and $z = 2.8-3.25$ mm). These laminae are
 297 characterized by an area fraction of cement and clays lower than 50 % with a near zero
 298 content of organics and fractures. The detrital quartz grains represent 40 to 50 % of area, and
 299 heavy minerals (mostly pyrite), micas and calcite are on the order of a few percent. The
 300 proportion of feldspars and ankerite is around 8-10 %.

301 **(2) Organic-rich lamina** ($z = 1.4-2.8$ mm). This lamina is 1.4 mm thick, and composed of
302 60 to 70 % of area as cement and clays with a higher organic and fracture content between
303 1.7 to 7 %. The proportion of detrital quartz is between 10 and 30 %. The heavy mineral
304 content is also higher than in the organic-matter poor lamina with variable proportions
305 between 2 and 5 %. Feldspars and ankerite are less common with area fractions between 2
306 and 6 %. Calcite and micas are still scarce, although peaks of micas and calcite were detected
307 due to very large micas at $z = 1.2$ mm and the presence of an horizontal calcite vein at $z =$
308 2.7 mm.

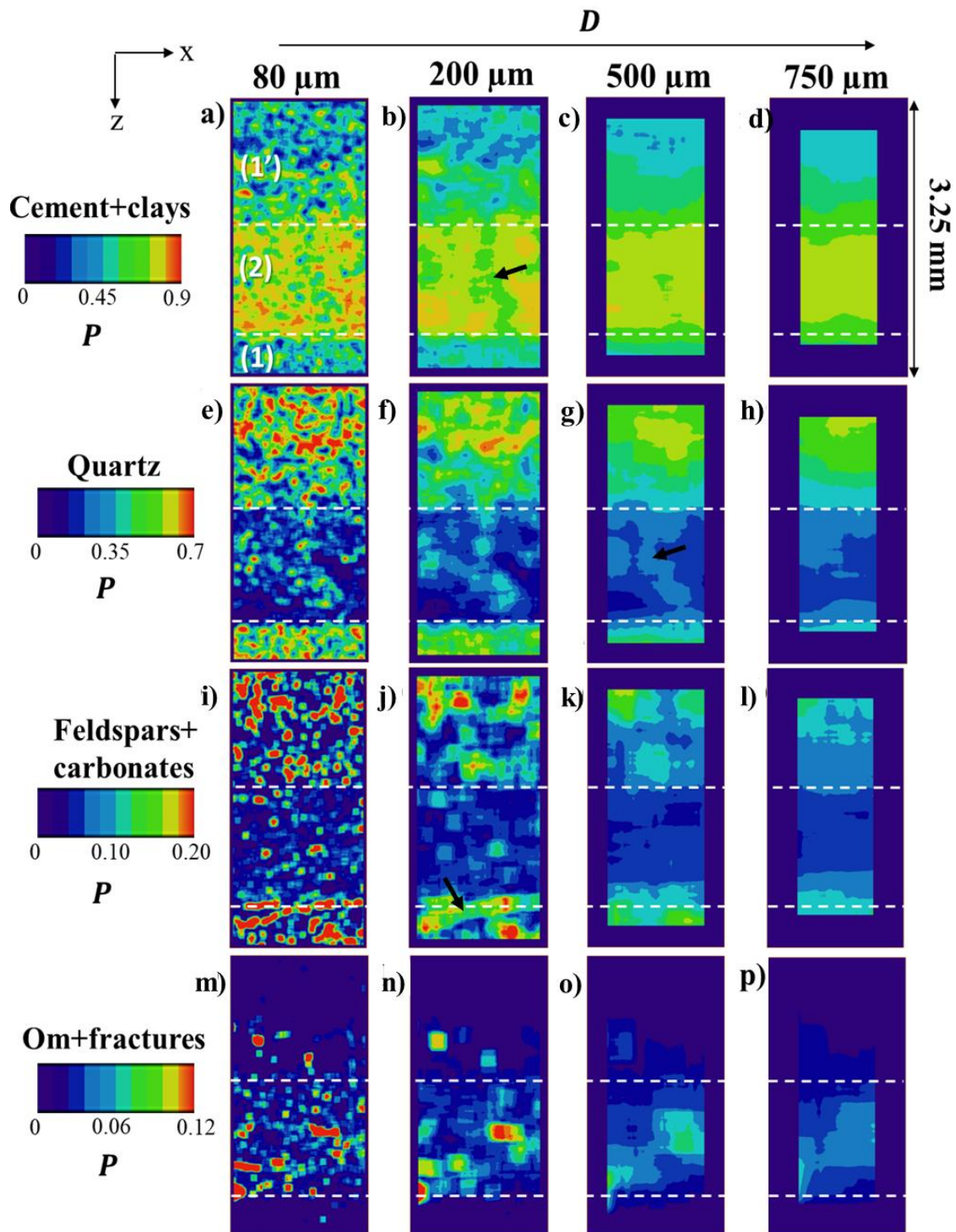
309 **Transition (1)-(2):** This is the transition between organic-poor and organic-rich laminae
310 ($z=0.80-1.4$ mm). It is marked by a progressive increase in cement and clay area content
311 from 40 to 62 %, and a decreasing proportion of detrital quartz grains from 50 to 30 %.
312 Feldspars/ankerite variability is less marked at between 6 and 2 %. The proportion of heavy
313 minerals increases from 2 to 5 %. However, the transition between the organic-poor and the
314 organic-rich laminae around $z = 2.8$ mm is very abrupt with sharp jumps in cement/clays,
315 organic-matter/fractures, quartz and heavy mineral proportions. The proportion of
316 cement/clays falls from 70 to 38 % over 240 μm (between $z = 2.68$ to 2.92 mm), while quartz
317 content dramatically increases from 9 to 49 % in 220 μm (between $z=2.76$ to 2.92 mm).

318 In order to ensure a sufficient dataset for statistics in sections 4.3 to 4.5 below, the organic-
319 poor and one part of the transition ($z = 0-1.28$ mm) zones were taken together to form a
320 gradual organic-poor lamina (1'), in order to compare its microstructure with the that of the
321 organic-rich lamina (2) ($z = 1.48-2.76$ mm).

322 **4.2. Spatial variability of phase content**

323 2D maps of P are shown as Fig. 4 for cement and clays, quartz, feldspars and carbonates, and
324 organics and fractures for $N=200, 500, 1250$ and 1876 pixels, i.e. for, 80, 200, 500 and 750

325 μm respectively (Fig. 4). $P = 1$ means that the domain D consists 100 % of the phase
 326 considered whereas 0 (purple) means it is completely absent.



327

328 **Fig. 4. 2D maps of phase area fractions with variable domain sizes for: (a-d)**
 329 **cement+clays, (e-h) quartz, (i-l) feldspars+carbonates, (m-p) Om+fractures. [Each of**

330 **the three laminae [(1) organic-poor lamina, (1') gradual organic-poor lamina, (2)**
331 **organic-rich lamina] are labelled on (a).**

332 The organic-rich lamina 2 is clay and cement-rich, and detrital quartz, feldspar and
333 carbonate-poor (Fig. 4 a-l). The organic-matter and fractures are heterogeneously distributed
334 due to the variable sizes of particles and fractures (Fig. 4 m-p). The organics and fractures are
335 nearly absent in the organic-poor lamina (1) (Fig. 4 m-p). The gradual organic-poor lamina 1'
336 is quartz, feldspar and carbonate-rich (Fig. 4 e-l). The heterogeneities relative to P are
337 defined by local higher or lower phase area fraction than the mean phase area fraction of each
338 map (local concentration of red or blue colors for example). For all phases and D sizes, the
339 area fraction is spatially heterogeneous in x (horizontal) and z (vertical) directions on the
340 whole mosaic, mainly due to the presence of a sedimentary lamination (Fig. 2a), but also to a
341 heterogeneous distribution of mineral phases inside each lamina.

342 For $D = 80 \mu\text{m}$, local high concentrations of quartz, feldspars, carbonates and organics (red
343 areas in Fig. 4 e,i,m, first column) defined the grain shapes visible on the mosaic in Fig. 2.
344 The phase heterogeneities are thus relative to the grain-size scale for $D = 80 \mu\text{m}$, due to the
345 small value of D including a low number of grains. Therefore this scale shows interfaces
346 between relatively large grains and the phase including cement and clays. For $D = 200 \mu\text{m}$,
347 the organic-rich lamina contains circular clay and cement-rich areas and a discontinuous
348 vertical line with a lower concentration (Fig. 4b), mainly due to the presence of quartz (Fig.
349 4f). Scattered feldspar and carbonate-rich areas were detected in the gradual organic-poor
350 lamina with a concentration gap around 0.15-0.20 with respect to the background. However,
351 the organic-poor lamina contains a continuous line with a higher concentration of feldspars
352 and carbonates ($P > 0.15$), mainly due to the presence of calcite veining (black arrow in Fig.
353 4j). This scale highlights the areas rich in large particles of organics and large fractures in the
354 organic-rich lamina (Fig. 4n). The maps show these interfaces between clusters and grain-

355 poor areas with the background at the scale of $D = 200 \mu\text{m}$. For $D = 500 \mu\text{m}$, cement and
356 clay proportion heterogeneities are averaged, reducing their surface fractions in both laminae
357 (Fig. 4c), whereas larger clusters of feldspars, carbonates, and quartz are highlighted (Fig. 4
358 g, k). A continuous vertical line of quartz ($P > 0.25$) is present in the organic-rich lamina
359 (black arrow in Fig. 4g). The organics and fractures are divided in two separate clusters with
360 $P > 0.06$ in the organic-rich lamina (Fig. 4o). For $D = 500 \mu\text{m}$, the domains D contain thus
361 more grains and average the phase area fraction and their heterogeneities relative to
362 individual and clusters of grains at larger scales than $D < 500 \mu\text{m}$. For $D = 750 \mu\text{m}$, phase
363 area fraction maps show the progressive and abrupt transitions between the different laminae
364 (Fig. 4 d, h, l, p, last column). They highlight the interfaces between sedimentary laminae,
365 and the maps for $D = 500$ show heterogeneities for extended clusters and the beginning of the
366 detection of laminae at an intermediate scale between $D = 200$ and $750 \mu\text{m}$.

367 The multi-scale mapping of phase area fractions shows three different types of
368 microstructural heterogeneities relative to three different scales: (i) the interfaces between
369 rigid inclusions (quartz, feldspars, carbonates, organics) and the phase cement/clays, (ii) the
370 interfaces between clusters of rigid inclusions and the background and (iii) the interfaces
371 between laminae. These maps highlight the high multi-scale variability of microstructure,
372 based on the area fraction parameter.

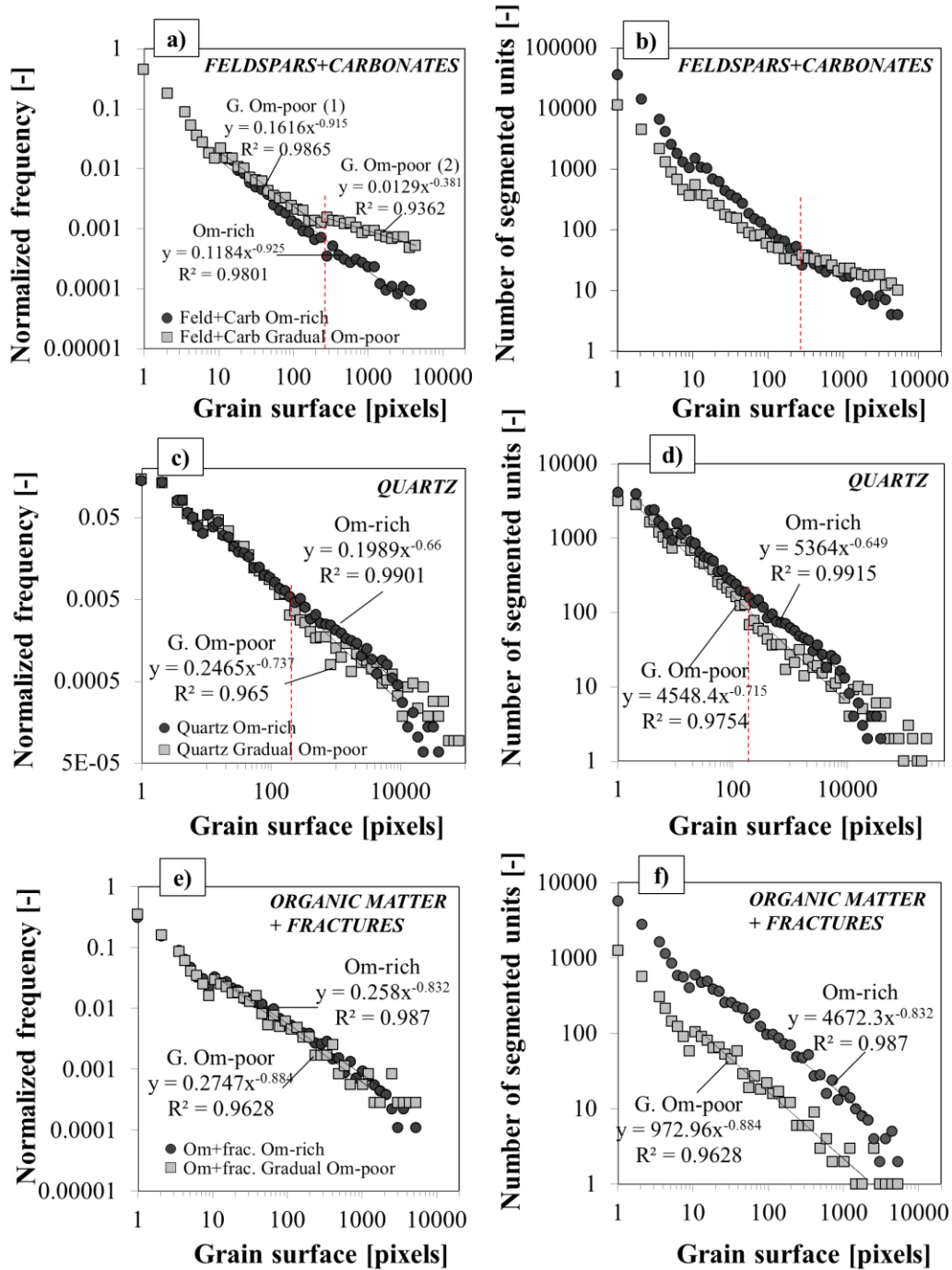
373 **4.3. Variability of microstructural parameters**

374 The grain size, grain number per size, length ratio and orientation of grains were calculated
375 for each of laminae 1' and 2 (labelled on Fig. 4a) in independent areas of 4204×3200 pixels
376 ($1.682 \times 1.280 \text{ mm}^2$) (Fig. 5).

377 *Variability of grain size*

378 The grain size distribution (GSD) is an important parameter to help understand the
379 sedimentation conditions. As expected in shale, the grain size distributions (Fig. 5 a, b, c),
380 follow a power-law for both laminae for quartz, feldspars, carbonates, organic matter and
381 fractures, micas and heavy minerals (not shown). Their fractal dimensions on the studied
382 areas are shown. In both laminae 1' and 2, the GSD of feldspars, carbonates and quartz are
383 similar for grain surfaces below 200 pixels (Fig 5 a, c) and differ significantly above 200 and
384 300 pixels respectively, especially for feldspars and carbonates (Fig. 5a). This difference is
385 interpreted as a result of a large proportion of large grains (surface above 200 pixels) of
386 feldspars and carbonates in the organic-poor lamina (Fig. 5b) alongside the presence of grain
387 clusters (aggregates of grains, red grains in Fig. 2a). The GSD for feldspars and carbonates
388 describes a dual power law, thus large grains and clusters contribute heavily to the total phase
389 proportion of the gradual organic-poor lamina. Quartz GSD differ for grains larger than 200
390 pixels but the effect is less pronounced than in feldspars and carbonates (Fig. 5c), whereas
391 quartz clusters are more easily visible in Fig. 2 (blue grains). The number of independent
392 quartz segments is fewer by 18 % in the gradual organic-poor lamina 1' than the organic-rich
393 lamina 2 (Fig. 5d), despite Fig. 2a suggesting the opposite. This can be explained by the fact
394 that the quartz clusters are significantly larger than the feldspar and carbonate clusters, hence
395 their number and frequency are consequently much lower. In the gradual organic-poor lamina
396 1', quartz regions are relatively small and independent, whereas they are larger and arranged
397 in clusters in the organic-rich lamina 2. The clusters decrease the number of individual quartz
398 segments, which minimize their impact on the GSD (Fig. 5 c,d).

399 Organic matter and fractures show very close GSD for both laminae 1' and 2 (Fig. 5 e) but
400 the number of segmented features is until 4.8 times higher in the organic-rich lamina 2 than
401 the gradual organic-poor lamina 1' (Fig. 5 d).



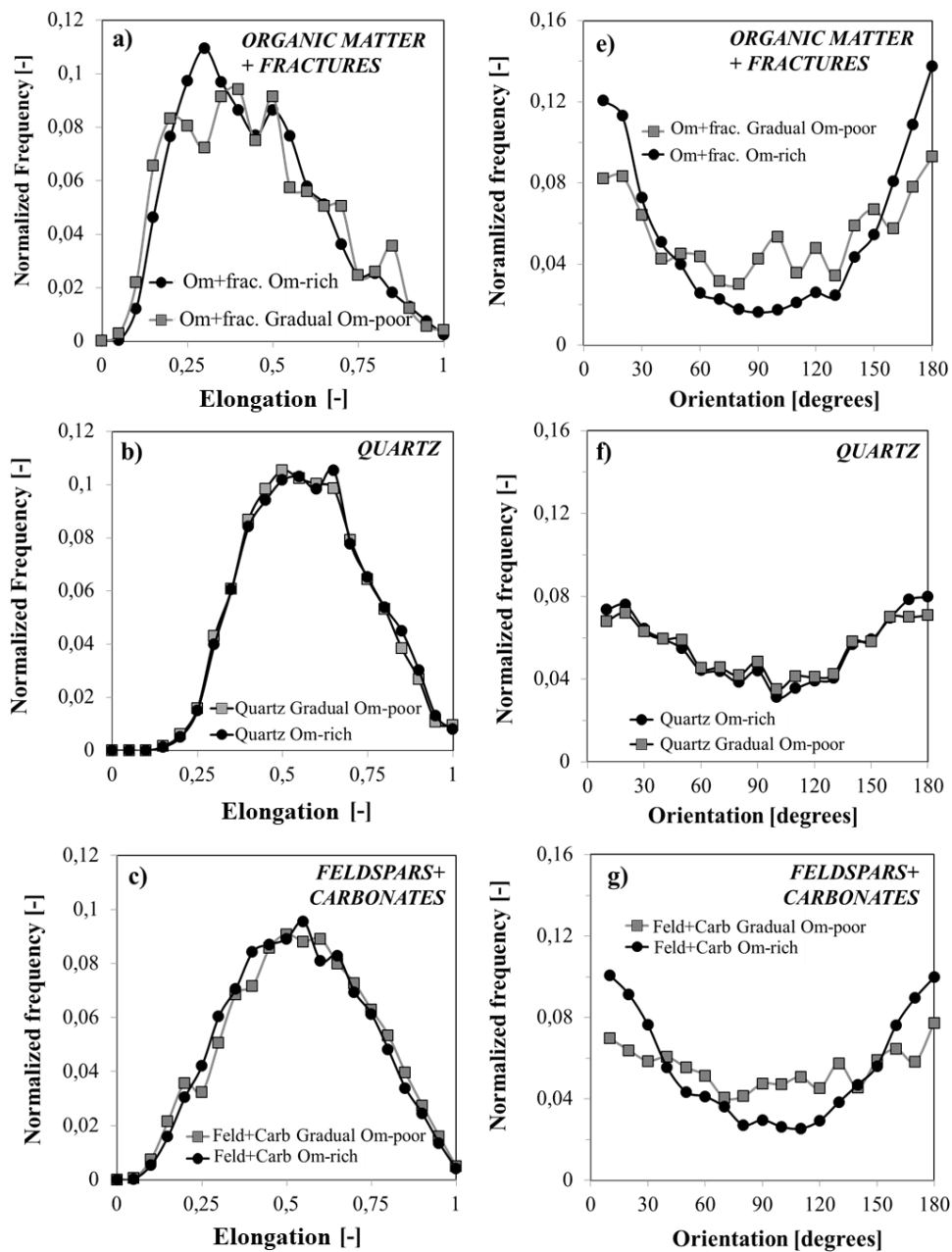
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403

Fig. 5. Grain size and grain number distributions for each of laminae 1' (gradual om-

404

poor) and 2 (om-rich).



406

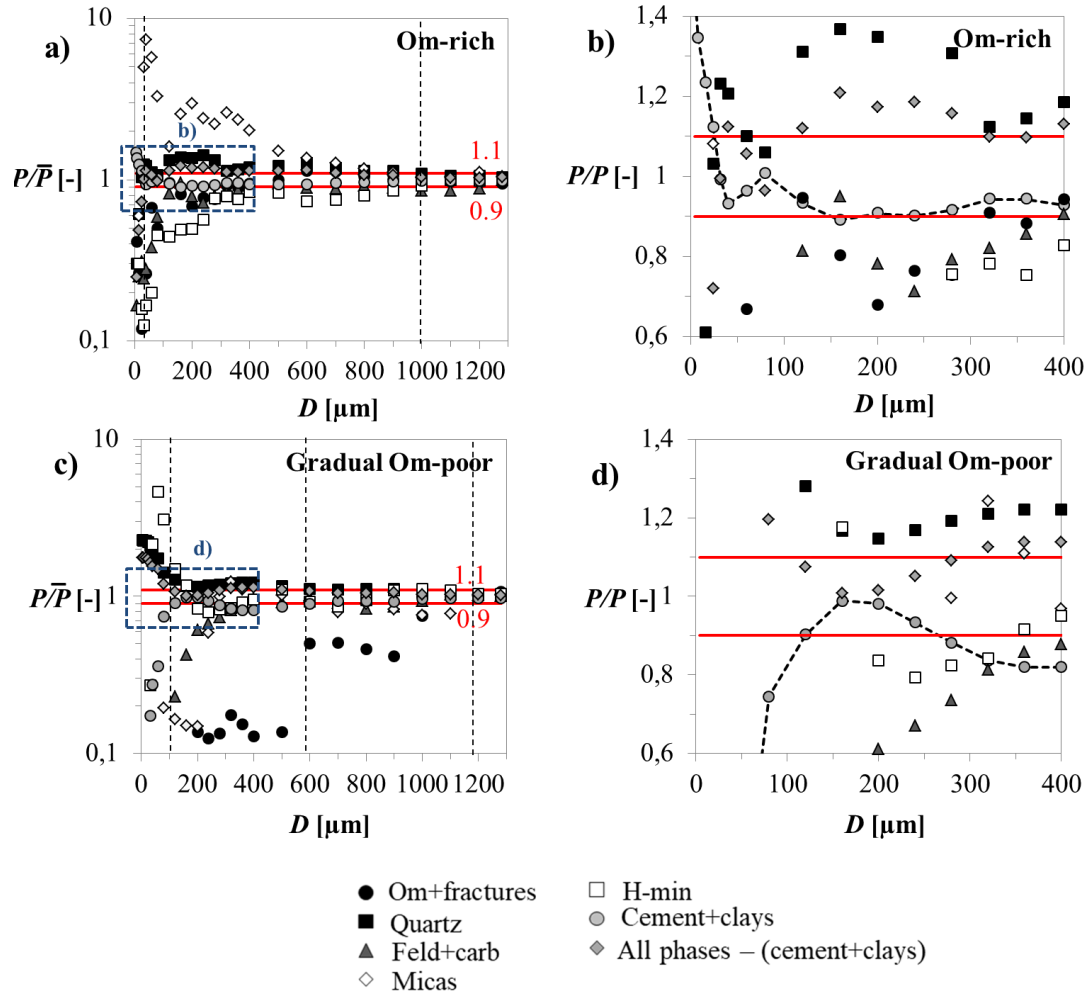
407 **Fig. 6. Elongation and orientation distributions for a), b) organic matter particles and**
 408 **fractures, c), d) quartz, e), f) feldspars and carbonates, on the organic-rich (black plots)**
 409 **and the gradual organic-poor (gray plots) laminae 2 and 1' respectively. Only grains**
 410 **larger than 10 pixels were considered.**

411 For both laminae, the microstructure of the Bowland Shale sample is anisotropic due to
412 horizontally-elongated quartz, feldspars, carbonates, organic matter particles and fractures
413 (Fig 6 a-g). The elongation distributions of quartz, feldspars, carbonates, organics and
414 fractures do not change significantly between the gradual organic-poor and the organic-rich
415 laminae (Fig. 6 a,b,c). However, the orientation distribution of organics, fractures, feldspars
416 and carbonates differ between the organic-rich lamina to the gradual organic-poor lamina
417 (Fig. 6 e,g). Organics, fractures, feldspars and carbonates are more elongated horizontally
418 (long-axis highest orientations at 0 and 180 degrees, i.e. parallel to the trace of bedding) in
419 the organic-rich lamina contrary to detrital quartz minerals which do not show any variation
420 of orientation between the two laminae (Fig. 6f). Based on these two criteria, the variation of
421 anisotropy is mainly due to a variation of orientation of fractures and feldspars, carbonates,
422 organic matter particles and fractures. Note that this measure of anisotropy ignores
423 contributions from any preferred crystallographic orientation of grains (other than micas) that
424 might exist and oriented cracks smaller than microscopic resolution.

425 **4.4. Quantitative description of the microstructure**

426 Figs. 3 to 6 have shown that the microstructure is spatially variable. However, even if the
427 microstructure is variable, microstructure modelling is required to predict strain localisation
428 and potential sites of fracture initiation in rocks (Lan et al., 2012; van den Eijnden et al.,
429 2016). In this section, three methods are used to quantify representative elementary areas of
430 the measured microstructure, based on the surface proportion of phases, and as applied to
431 polycrystalline and heterogeneous materials (Kanit et al., 2006; Madi et al., 2005) and rocks
432 (Klaver et al., 2012; Houben et al., 2014; Vik et al., 2014). These methods can quantify how
433 representative are the microstructural models generated from the microstructural parameters
434 of the mosaic.

435 ***Counting-box approach***



436

437 **Fig.7. (a and b) Ratio between the local phase area fraction P and the mean phase area**
 438 **fraction \bar{P} as a function of the size of calculation domain D for the *organic-rich lamina***
 439 **where b) is a magnified view of the dashed-line box in a) for $D \leq 400 \mu\text{m}$. (c and d) show**
 440 **similar features for the *gradual organic-poor lamina*, where d) is a magnified view of the**
 441 **dashed-line box in c) for $D \leq 400 \mu\text{m}$.**

442 The starting point of the counting box method is the center of each lamina image, in order to
 443 take into account the maximum number of different D values. P is the mean area fraction of
 444 the phase in D and \bar{P} is the mean phase area fraction in the whole image of the lamina. In the
 445 organic-rich lamina, the ratios P/\bar{P} for cement-clays (gray circles in Fig. 7 a,b) and all rigid
 446 inclusions (gray diamonds in Fig. 7a,b) start to oscillate at 1 ± 0.10 for D close to $50 \mu\text{m}$ and

447 320 μm , respectively. However, the ratio P/\bar{P} of all phases separately considered is stable at
 448 1 ± 0.10 for $D \geq 1$ mm. This result is interpreted as being a balance effect between the
 449 different mineral phase fractions. In the gradual organic-poor lamina 1' (Fig. 7 c, d), P/\bar{P}
 450 varies at 1 ± 0.10 for $D \geq 120$ μm for the cement-clays (oscillation around 0.9) and the overall
 451 grain phase (oscillation around 1.1). However, all phase area fractions taken independently
 452 are stable for $D \geq 600$ μm , except organics and fractures at $D \geq 1.1$ mm due to small and
 453 elongated units (Fig. 5f, 6d) and a low mean area fraction on the whole image. Consequently,
 454 P/\bar{P} reaches 1 ± 0.10 for $D \geq 1.2$ mm for all phases separately considered. The
 455 homogenization areas are larger for the gradual organic-poor lamina than the organic-rich
 456 lamina due to higher grain sizes and larger clusters, and the scattering of small and low
 457 frequency phases such as organics and fractures.

458 *Dispersion of phase proportion*

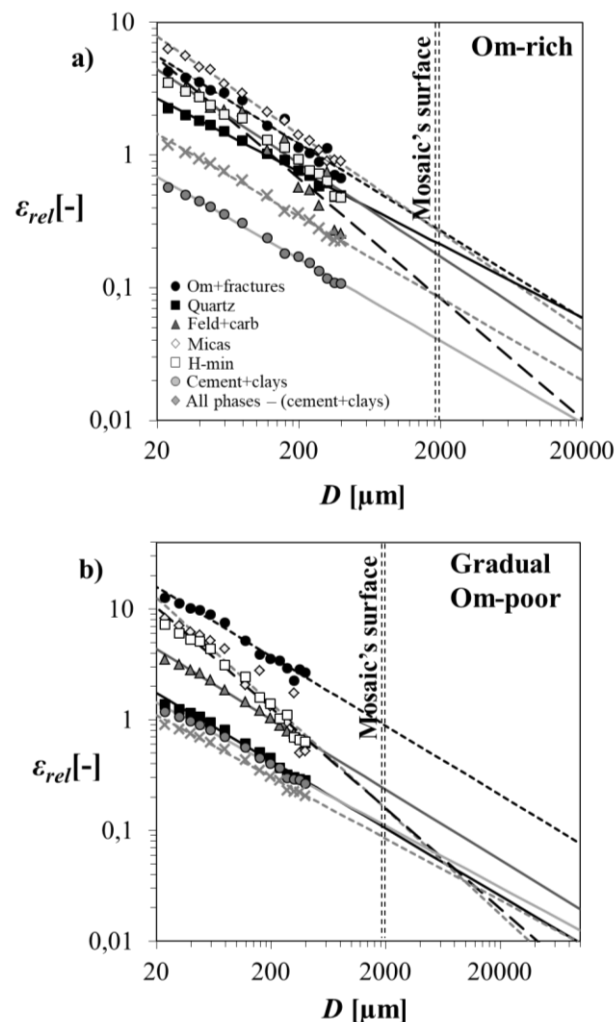
459 Each lamina was divided into a number, N , of independent domains, D , to calculate the
 460 standard deviation $\sigma_P(D)$, variance $\sigma_P^2(D)$ and relative error $\varepsilon_{rel}(D)$ (Fig. 8) of each phase
 461 area fraction P as a function of D (Kanit et al., 2003) (Equation 5, Fig. 8, Tables 1,2). :

$$\varepsilon_{rel}(D) = \frac{2\sigma_P(D)}{\bar{P}\sqrt{N}} \quad (5)$$

462 with \bar{P} the mean phase area fraction between 0 and 1.

463 In Kanit et al., (2003), N is the number of cells on theoretical Voronoï mosaics. In this study,
 464 N is the number of cells, called domains, of the experimental mosaic. A domain can therefore
 465 be considered as a 2D rock subsample of the whole mosaic of the Bowland Shale. The
 466 experimental mosaic has a limited size, so the number of domains is also limited to a finite
 467 value.

468 The fitting of relative uncertainty ε_{rel} by a power law $\varepsilon_{rel} = a.D^{-b}$ (Table 1) allows the
 469 quantification of ε_{rel} for the surface corresponding to each type of lamina (Fig. 8, table 1).
 470 On each lamina, the number of calculation domains as a function of the domain's size follows
 471 the law: $N = 2.10^6 . D^{-b}$ with $b = 2.05$ ($r^2 = 0.9993$) for D between 10 and 1250 pixels (4 and
 472 500 μm respectively). Data with fewer than 10 domains were not considered (Kanit et al.,
 473 2003), i.e domains above 400 μm . As expected, the dispersion of P decreases when the size
 474 of D increases for phases segmented. The relative errors of all phase area fractions were
 475 calculated as a function of D for both laminae according to equation 5 (see Figure 8).



476

477 **Fig. 8. Relative uncertainty, ε_{rel} , of area fraction as a function of D for a) the organic-**
 478 **rich lamina and b) the gradual organic-poor lamina, for $N=1$.**

479 Considering all phases combined together in both laminae, $\varepsilon_{rel} = 0.11$ and 0.09 for square
 480 domains of $1.32 \times 1.32 \text{ mm}^2$ and $1.76 \times 1.76 \text{ mm}^2$ respectively (gray crosses in Fig. 8),
 481 whereas $\varepsilon_{rel} \gg 0.1$ for the phases segmented independently for same D . Based on the surface
 482 proportion, the areas used to characterize the lamina's microstructures are representative with
 483 a ε_{rel} around 10% when all mineral phases are considered together in a unique phase, but not
 484 separately, at the study resolution.

<i>Gradual Om-poor</i>	a	b	R²
Om+fractures	103	0.626	0.97
All grains	5.80	0.556	0.99
Cement+clays	7.54	0.556	0.99
Quartz	10.8	0.610	0.99
Feldpars+Carbonates	29.1	0.635	0.99
Heavy minerals	154	0.906	0.98
Micas	218	0.952	0.90
<i>Om-rich</i>			
Om+fractures	39.8	0.658	0.95
All grains	9.19	0.619	0.99
Cement+clays	4.39	0.619	0.99
Quartz	13.9	0.551	0.99
Feldpars+Carbonates	81.9	0.908	0.91
Heavy minerals	36.6	0.706	0.99
Micas	71.3	0.738	0.99

485

486 **Table 1. Parameters of the power law fitting, ε_{rel} , as a function of D corresponding to**
 487 **both laminae.**

		REA [mm]								
		N=1			N=5			N=10		
		ϵ_{rel}	0.15	0.1	0.05	0.15	0.1	0.05	0.15	0.1
Organic-rich	Cement+clays	0.233	0.449	1.38	0.063	0.122	0.375	0.036	0.070	0.214
	Grains	0.746	1.44	4.40	0.203	0.391	1.20	0.116	0.223	0.685
	Quartz	3.71	7.75	27.2	0.861	1.80	6.33	0.459	0.959	3.37
	Feld+Carb	1.03	1.62	20.4	0.426	0.666	1.43	0.291	0.455	0.975
	H-min	2.41	4.28	11.4	0.771	1.37	3.65	0.472	0.838	2.24
	Om+fractures	4.83	8.94	25.6	1.42	2.63	7.55	0.839	1.55	4.46
	Micas	4.24	7.35	18.8	1.42	2.47	6.32	0.892	1.54	3.95
Gradual Organic-poor	Cement+clays	1.15	2.38	8.28	0.270	0.560	1.95	0.145	0.300	1.04
	Grains	0.715	1.482	5.16	0.168	0.349	1.21	0.090	0.187	0.650
	Quartz	1.10	2.14	6.68	0.295	0.573	1.79	0.167	0.325	1.01
	Feld+Carb	4.02	7.61	22.7	1.13	2.14	6.38	0.655	1.24	3.70
	H-min	2.12	3.32	7.14	0.874	1.37	2.94	0.596	0.932	2.03
	Om+fractures	33.8	64.7	196	9.36	17.9	54.1	5.38	10.3	31.1
	Micas	2.10	3.21	6.66	0.901	1.38	2.86	0.626	0.959	1.97

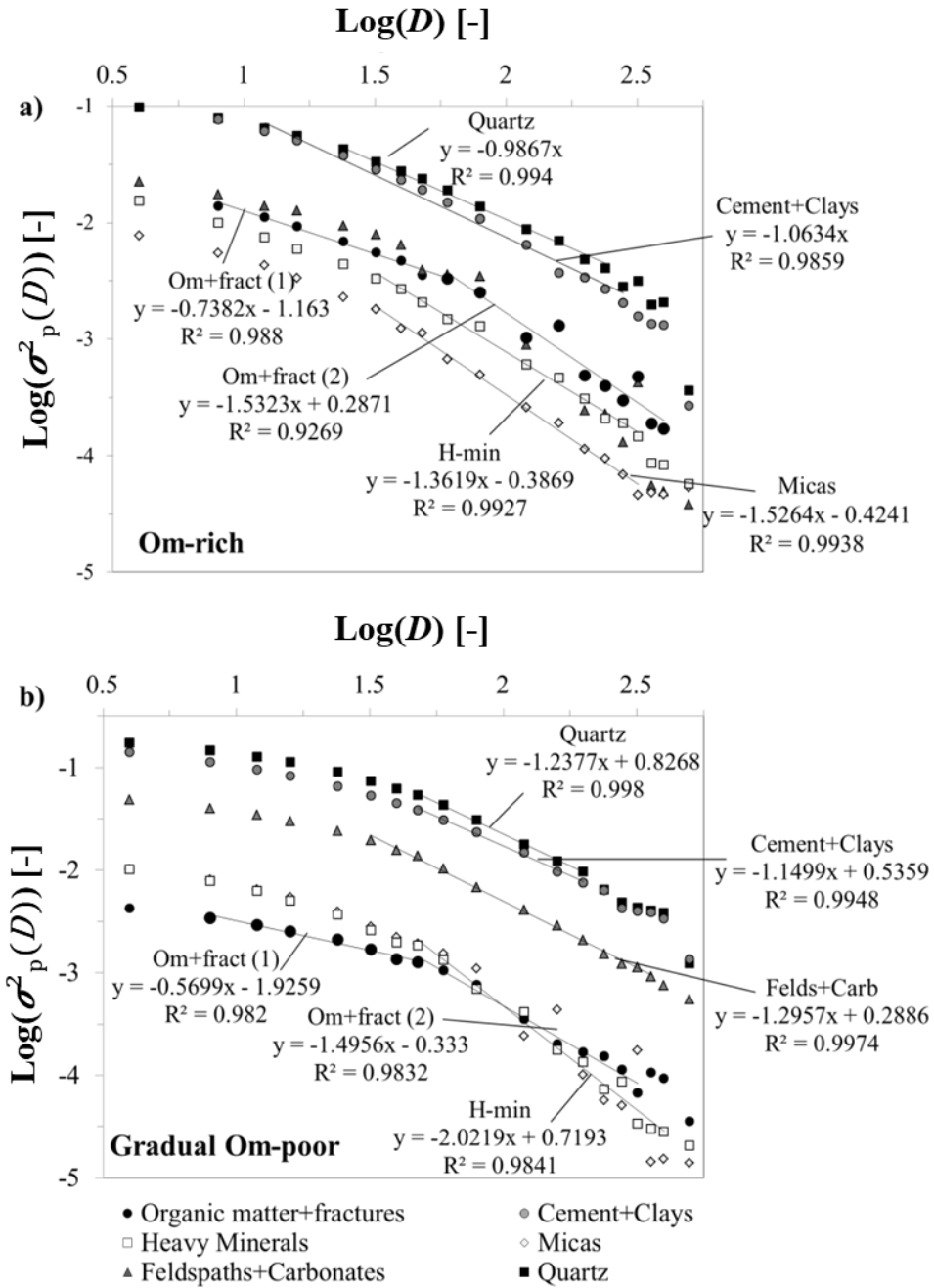
488

489 **Table 2. REA values in mm, calculated for different number of domains and for the**
490 **phases of each lamina.**

491 REA were calculated for a number of samples of 1, 5 and 10 and for $\epsilon_{rel} = 0.05, 0.1$ and 0.15
492 (Table 2) according to Table 1 and Fig. 8. For $\epsilon_{rel} \ll 0.05$, unrealistically high values of
493 REA were found, significantly greater than lamina width. Therefore, these values were not
494 considered to be realistic. The REA calculated for $\epsilon_{rel} = 0.1$ and $N = 1$ are millimetric
495 surfaces. However, the maximum possible lamina width used to calculate the microstructural
496 parameters is only 1.5 mm due to natural sedimentary structure (Fig 2). As such, the
497 calculations for $N = 10$ and $\epsilon_{rel} = 0.1$ are more realistic than for $N = 1$ or 5 because the REA
498 are below 1.5 mm for all phases, except organics and fractures in the gradual organic-matter
499 poor lamina owing to their small quantities and scattered structure.

500 ***Randomness of phase distribution***

501 The variance of phase area fraction describes a log-linear law as a function of D for both
502 laminae (Fig. 9, equation 6).



503

504 **Fig. 9. Fitting of $\text{Log}(\sigma_p^2(D))$ as a function of $\text{Log}(D)$ for the mineral phases of a) the**
 505 **organic-rich lamina and b) the gradual organic-poor lamina.**

$$\text{Log}(\sigma_p^2) = -\alpha \text{Log}(D) + k$$

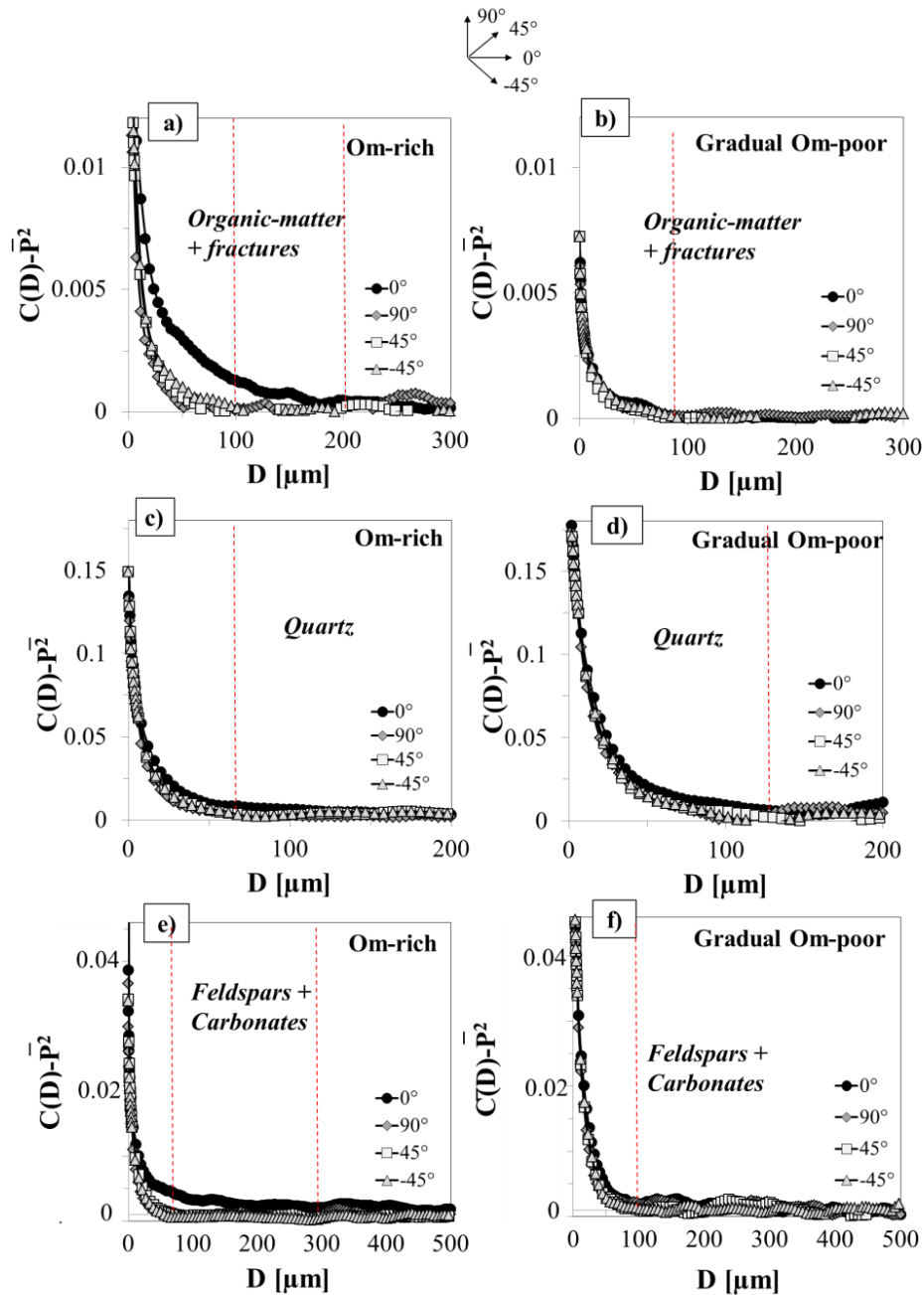
(6)

506 α in Equation 6 is an indicator of the randomness of the distribution of the physical parameter
507 studied, which is P in this study (Cailletaud et al., 1994; Kanit et al., 2003;). According to
508 these authors, P was considered to be randomly distributed for $\alpha = 1 \pm 0.15$. $\alpha > 1$ means that
509 the variance increases rapidly with increasing domain, D , whereas $\alpha < 1$ when the variance
510 decreases with increasing domain, D .

511 In the organic-rich lamina, the proportions of quartz and cement-clays are distributed
512 randomly enough for $D = 24$ to $400 \mu\text{m}$ ($\log(D)=1.38$ to 2.6) and $D = 12$ to $400 \mu\text{m}$
513 ($\log(D)=1$ to 2.6) respectively, unlike the other phases (Fig. 9a). In the gradual organic-poor
514 lamina, only the fraction of cement-clays is randomly distributed for $D = 48$ to $200 \mu\text{m}$
515 ($\log(D)=1.68$ to 2.3) whereas all other phases are heterogeneously scattered, with $\alpha \gg 1$ for
516 a domain larger than $48 \mu\text{m}$. For all phases, the coefficients α are higher in the gradual
517 organic-matter poor lamina than in the organic-matter rich lamina. Therefore, the variance of
518 their fractions increases more rapidly with increasing D in the gradual organic-poor lamina
519 than in the organic-rich lamina. The gradual organic-matter poor lamina contains more large
520 grains and clusters (especially quartz, feldspars and carbonates), and consequently, their
521 number per domain is lower than if they were smaller grains. It thus requires a larger domain,
522 D , to generate a homogeneous phase area fraction on a finite surface. In the whole range of
523 D , $\log(\sigma^2)$ of area fraction for all phases show affine laws in both laminae, especially visible
524 in the gradual-organic matter poor lamina. An example is given for the organics and fractures
525 (Fig. 9 a, b). For organics and fractures, $\alpha \ll 1$ for $D \leq 60 \mu\text{m}$ ($\log(D)=1.78$), while $\alpha \gg 1$
526 for $D > 60 \mu\text{m}$. For $D \leq 60 \mu\text{m}$, the resolution is not sufficient to capture the different sizes of
527 organics and fractures. However, domains larger than $60 \mu\text{m}$ include various sizes of particles
528 heterogeneously scattered and/or with insufficient mean fraction to cause a sharp increase of
529 variance as a function of D . The log-linear law is divided in two slopes, caused by a scaling
530 effect due to various particle sizes, but a fixed resolution.

531 *Covariance*

532 In order to compare the different covariograms, they are plotted minus the square of the mean
 533 phase area fraction in each lamina in Figure 10.



534

535 **Fig. 10. Covariograms of a) organic-matter and fractures in the Om-rich lamina; b)**
 536 **organic-matter and fractures in the gradual Om-poor lamina; c) quartz in the Om-rich**

537 **lamina; d) quartz in the gradual Om-poor lamina; e) feldspars and carbonates in the**
538 **gradual Om-poor lamina; and f) feldspars and carbonates in the Om-rich lamina.**

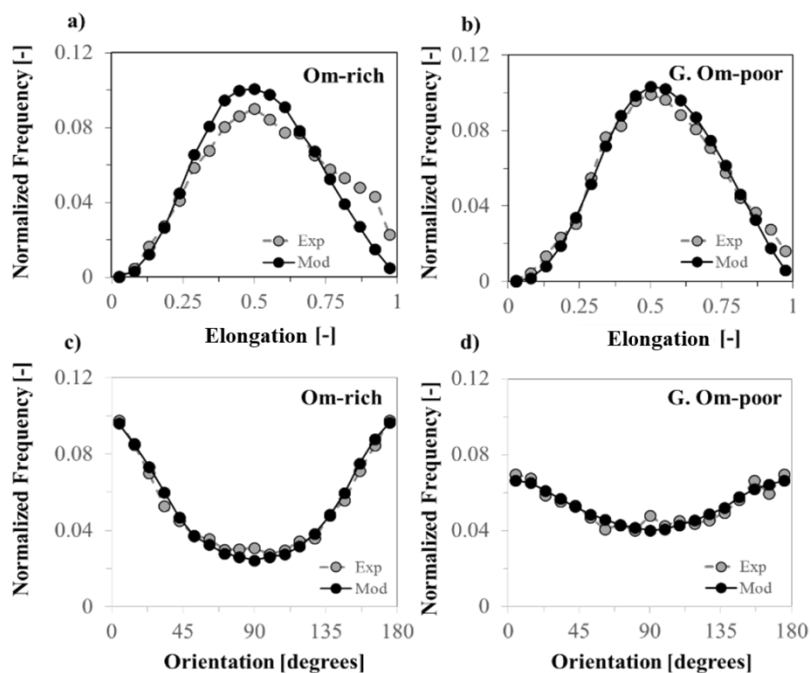
539 Organic-matter and fractures become homogeneously scattered for $D \geq 100 \mu\text{m}$, except in the
540 0° orientation with $D \geq 200 \mu\text{m}$, due to horizontal fractures and elongated particles of
541 organics in the organic-matter rich lamina (Fig. 10a, b). Under the study conditions, domains
542 smaller than $100 \mu\text{m}$ can be considered as heterogeneous scales for the organic-poor lamina,
543 and $200 \mu\text{m}$ for the organic-rich lamina. The covariograms of quartz reach the asymptote for
544 domains $D \geq 75 \mu\text{m}$ in the organic-matter rich lamina and $120 \mu\text{m}$ in the gradual organic-
545 poor lamina (Fig. 10 c, e). The higher heterogeneous scale relative to quartz in the gradual
546 organic-poor lamina is mainly due to the presence of aggregates which contribute towards an
547 increase in the size of quartz heterogeneities.

548 In the gradual organic-poor lamina, the covariograms of feldspars and carbonate reach the
549 asymptote for D close to $100 \mu\text{m}$ in all directions, showing a microstructure close to isotropic
550 for these phases at the resolution of the study. However, this is not the case for the organic-
551 rich lamina. The asymptote is reached for $D=300 \mu\text{m}$ in the horizontal direction (0°) contrary
552 to the other directions (asymptotes at $75 \mu\text{m}$), due to smaller, more elongate and horizontally-
553 elongate feldspars, as well as horizontal calcite grains within a vein in the organic-rich lamina
554 (bottom part of the lamina, Fig. 2). Therefore, the laminae have different scales of textural
555 heterogeneities. For the microstructure models, the parameters are calculated over a surface
556 of $1.32 \times 1.76 \text{ mm}^2$; at least 3 to 11 times the size of the minimal homogeneous surfaces
557 determined by the covariograms (considering all directions). In the sample of Lower
558 Bowland Shale used here, the variability of microstructure thus causes a variability of the
559 minimum size of the homogeneous scale. Covariograms also indicate that the anisotropy of
560 the microstructure is mainly caused by the organic-matter particles, fractures, feldspars and

561 carbonates in the organic-rich lamina (Fig. 10 a, c, e). The microstructure is more isotropic in
 562 the gradual organic-poor lamina (Fig. 10 b, d, f).

563 5. Microstructural modelling

564 The total grain geometry of the model was calibrated against the geometry of all mineral
 565 types combined. This calibration, performed against distributions of elongation ratio and
 566 orientation, leads to the total distribution of all phases in the model, including the ones that
 567 eventually make up the cement and clays. Results of this first step are given in Fig. 11. The
 568 overall properties are calibrated at $\xi = 1.3$ in the organic-rich lamina and $\xi = 1.1$ in the
 569 gradual organic-poor lamina. ξ is higher for the organic-rich lamina because of particles that
 570 are more horizontally-elongated in the organic-rich lamina which therefore cause a greater
 571 stretching of the modelled microstructure.



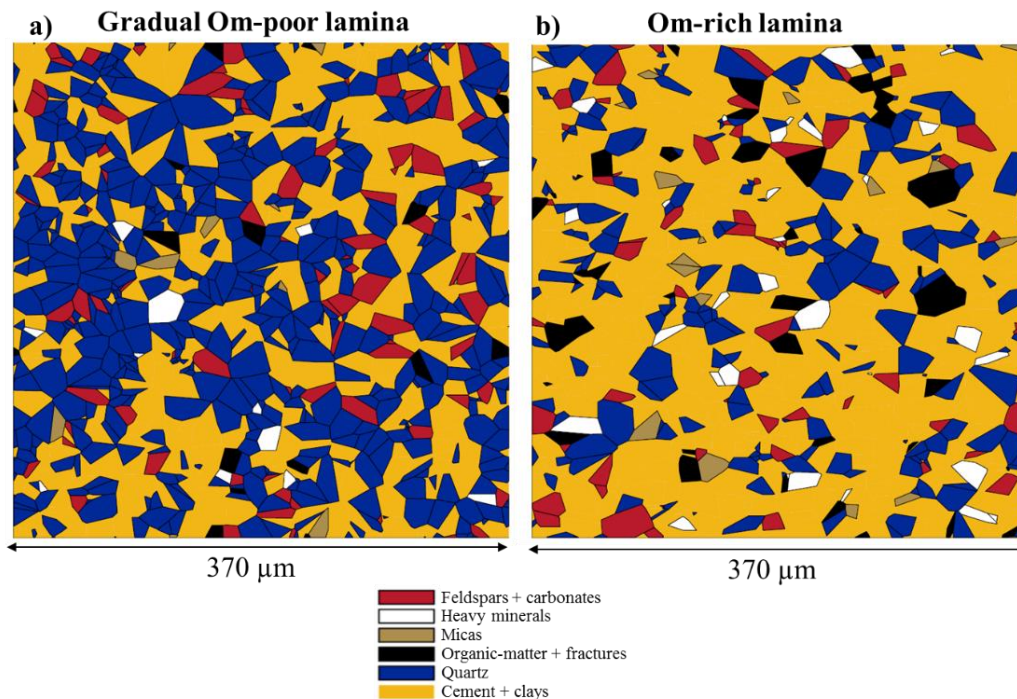
572

573 **Fig. 11. Result of the model calibration of all model grains against the data of all phase**
 574 **units combined [quartz + (feldspars,carbonates) + micas + heavy-minerals +**
 575 **(organics,fractures)]: a) elongation distributions of the organic-rich lamina; b)**

576 elongation distribution of the gradual organic-poor lamina; c) orientation distribution
577 of the organic-rich lamina; and d) orientation distribution of the gradual organic-poor
578 lamina.

579 The distributions for elongation and orientation of the microstructure model match well the
580 experimentally observed distributions in the case of all phases combined (Fig. 11). Only the
581 distribution of the elongation of the grains in the organic-rich lamina appears poorly
582 calibrated (Fig. 11a), but this can be corrected for in the model through proportionally
583 assigning mineral properties.

584 Figure 12 shows two resulting periodic microstructures, reproducing the distributions of
585 elongation, orientation and surface fractions of the gradual organic-matter poor lamina (Fig.
586 12a) and the organic-matter rich lamina (Fig. 12b). Other periodic microstructures are given
587 in Appendix 1 and the distributions of elongation ratio and orientation for each mineral type
588 are shown in Appendix 2.



589

590 **Fig. 12. Geometrical microstructure models of a) the gradual organic-poor lamina, b)**
591 **the organic-rich lamina. The models were generated from 1000 cells of all phases**
592 **combined.**

593 In a numerical sense, a scale bar should not be displayed with the microstructure model.
594 However, shales are multi-scale materials with components and properties varying over this
595 same range of scales. The scale is therefore important in order to determine which
596 phenomena can be studied with these models, because in shale materials phenomena
597 associated with one scale can exhibit controls over behaviors on larger scales. The models
598 presented here are at the mesoscopic scale, effectively micrometric rigid inclusions within a
599 “binder” composed of clays and quartz cement. A scale was qualitatively calculated for the
600 microstructure models in Fig. 12. Considering the mean cell size for each phase and 1000
601 cells in total, the scale value suggested and shown on Fig. 12 is around 370 μm for both
602 laminae.

603 **6. Discussion**

604 **6.1. Variability of interfaces relative to phase area fraction, from the μm to** 605 **mm scale**

606 Microstructural defects influence the initiation, propagation and aperture of fractures within
607 shales (Montes et al., 2002; Hedan et al., 2012; Fauchille et al., 2014, 2016; Wei et al., 2016).
608 The maps of phase area fractions highlight the location of three types of interface: the limits
609 between (1) individual grains and the cement and clay phases (micrometer scale), (2)
610 discontinuous phase aggregates and the background (several tens of micrometer scale), and
611 (3) the transitions between laminae (millimeter scale). These interfaces are detected as a
612 function of the size of the calculation domain, which is directly correlated to the scale

613 investigated. The capture of these different interfaces is only possible with a large mosaic of
614 images in high resolution, which allows a multi-scale investigation of the microstructure.
615 Multi-scale 2D mapping allows the detection of heterogeneities and interfaces which may not
616 be obvious when only examining at the initial mosaic, especially for aggregate boundaries.

617 The Lower Bowland Shale has a complex and multi-scale natural fracture network (Fauchille
618 et al., 2017). The Lower Bowland Shale sample studied here may have also an interesting
619 potential for “artificial” (such as hydraulic simulation) fracture propagation in multiple
620 directions, due to the variability of interfaces relative to the microstructure.

621 The transport properties of shales are controlled by the structure and arrangement of potential
622 transport pathways (Keller et al., 2011), particularly at the nanoscale (McKernan et al., 2017).
623 Microcrack location and properties are important for any bottom-up prediction of the
624 transport properties of shales. The quantification of strain concentrations at specific
625 interfaces using digital image or volume correlation may present an interesting way forward
626 in the development of predictive models of fracture initiation and propagation, and in the
627 understanding of scale factors controlling the fracture mechanisms.

628 **6.2. Contrasting vertical microstructural properties in the Lower Bowland** 629 **Shale, from the μm to mm scale**

630 According to previous studies, the qualitative variability of microtexture of Bowland Shale
631 from Preese-Hall-1 lies at the borehole scale with various microtexture types (Andrews,
632 2013; Fauchille et al., 2017). Here, we demonstrate quantitatively that it is also variable at the
633 sample scale, from the mm to the μm at the resolution used. The alternation of
634 mineralogically and texturally variable laminae indicates periodic changes of depositional
635 controls during the sedimentation process.

636 With different mineral content, grain properties and anisotropy, the laminae may have
637 markedly different mechanical properties and fracturing behavior, both within each layer, but
638 perhaps particularly at their transitions, which are smooth or abrupt, but necessarily
639 elastically contrasting. These different properties and behavior could influence the production
640 and orientation of microcracks that are likely to control permeability and contribute
641 significantly to anisotropy (McKernan et al., 2017). The organic-poor lamina is quartz-rich
642 (detrital quartz and cement), unlike the organic-rich lamina. A relatively low proportion of
643 clay minerals (<35%) (Andrews, 2013) and high proportion of non-swelling minerals is
644 considered to be favorable for hydraulic fracturing cutting across the layering (Jarvie, 2012,
645 2014), depending on the in-situ stress state, and hence for potential gas extraction. High
646 proportions of quartz or carbonates that are contiguous limit ductility and enhance the
647 brittleness of the rock (Jarvie 2014; Raji et al., 2015). Microtextural changes across layers
648 induce discontinuities and weak regions, producing localized stresses, and strain
649 incompatibilities under mechanical simulations (Lan et al., 2010 ; Amann et al., 2011) which
650 aid in the development of pathways for fracture growth.

651 However, the presence of a laminated structure can act to limit the vertical extent of fractures
652 by blunting cracks propagating across the layering. Ease of formation of shear steps parallel
653 to laminae further acts to inhibit vertical fracture growth by blocking upward flow paths for
654 hydraulic fracture fluid (Rutter and Mecklenburgh, 2017). The presence of rather thin
655 laminae increases the number of interfaces relative to thick beds, and thereby improves the
656 number of potential pathways for fracturing. At such larger scales, in the Austin Chalk for
657 example, the vertical connectivity of fractures is controlled by the vertical alternation of chalk
658 and shale layers due to different microstructure (grain size), fracture toughness and maximum
659 compressive stress (Warpinski and Teufel, 1989; Rijken and Cooke, 2001). According to
660 previous studies, the Bowland Shale contains laminated and interlaminated microtexture

661 types with laminae less than millimeter-thick. Note that the thickness of laminae is much
662 smaller and periodic in the interlaminated microtexture than the laminated microtexture.
663 Thinner shale layers have lower resistance to fracturing compared to thick beds (Rijken and
664 Cooke, 2001). The characterization of the mechanical behavior of each type of lamina will
665 aid understanding of the impact of the lamination on the vertical propagation of hydraulic
666 fractures in the Bowland Shale.

667 **6.3. Comparison of various methods for estimating the homogeneous scales** 668 **of the Bowland Shale microstructure**

669 The counting-box and the covariance methods are easier and faster to perform than the 2D
670 mapping and dispersion approaches of Kanit et al., (2003) which are more time-consuming
671 due to the requirement for incremental calculations on millions of pixels. Even if the center of
672 the first domain can be chosen randomly, the counting-box method gives a local
673 approximation of the representative area of the total surface investigated, for specific physical
674 parameters, scale and resolution. Nevertheless, the results depend strongly on the final
675 surface because the comparative surfaces overlap. For example, the domains of 1 and 1.2 mm
676 have an overlap of 83.3% ($100 \times 1/1.2 = 83.3\%$). As the calculation domain becomes closer
677 to the final surface (i.e. 1.76×1.32 mm in this case), the probability that the physical
678 parameter (area fraction here) measured on the subsurface is close to the mean parameter of
679 the entire surface increases and reaches 1.

680 In the dispersion approach, or mosaic scattering, the mosaic is divided into tens to several
681 thousands of independent domains which give a sufficient set of data for statistical
682 interpretation. The domains are compared to each other, rather than to a mean parameter
683 measured over the whole surface. However, the requirement for this substantial number of
684 subareas does not allow the use of areas larger than 400×400 μm in our case, whereas

685 calculations are performed for millimeter-scale areas in the counting-box and covariance
686 approaches. The dispersion approach therefore limits the maximum size of the subareas used.
687 The representative areas are thus determined by extrapolation using a power law and not
688 directly from experimental data.

689 For each phase unit considered independently from the others, the homogeneous surfaces of
690 phase area fraction are obtained at 1-1.2 mm with a relative error of 10% with the counting
691 box, whereas they are 7 to 9 mm with the dispersion approach. For all phase units combined,
692 the homogeneous areas are much smaller due to a balance effect caused by the mixing of low
693 proportion phases. REA are in the range of 150-350 μm with the counting box method,
694 whereas they are around 1.44 mm with the dispersion approach for all phases combined to
695 form a single phase. As per previously, the REA are up to 9 times larger with the dispersion
696 approach than with the counting box. The 2D quantitative maps (Figure 4, Appendix A)
697 confirm that the homogeneous surfaces are larger than $500 \times 500 \mu\text{m}$ for the organic-rich
698 lamina, and larger than $750 \times 750 \mu\text{m}$ for the organic-poor lamina, which support using the
699 dispersion approach to estimate the REA. The counting box was found to underestimate the
700 size of the representative elementary surface. The 2D mapping of phase area fraction
701 provides a direct method of viewing the parameter under consideration, therefore is useful in
702 the comparison of methods.

703 The homogeneous surfaces determined by the covariance approach are much smaller than
704 those obtained by the counting box and dispersion methods (Rolland du Roscat et al., 2007).
705 Indeed, the covariance is used to define homogeneity relative to the size of objects comprising
706 the image (Fig. 2b), i.e., the characteristic length of heterogeneities, in addition to their area
707 fraction. The size of the homogeneous scales determined by the dispersion approach appears
708 proportional to the number of heterogeneities, with a characteristic length determined by the
709 covariance (Rolland du Roscat et al., 2007).

710 Therefore, we recommend using the dispersion approach when the surface is large enough to
711 have several sizes of subareas, in order to extrapolate REAs at different relative errors. This
712 is supported by the 2D quantitative maps showing that the counting box and the covariance
713 approaches are not sufficient to define a homogeneous scale of the area fraction on a large
714 surface. The covariance gives a minimum size of homogeneous scale which is important in
715 the case that a larger surface cannot be investigated.

716 The counting-box and covariance methods are widely used in image analysis, and geoscience
717 generally. On the other hand, the dispersion method introduced by Kanit et al., (2003) has not
718 been very commonly applied in geoscience, and particularly to shales, except for recent
719 studies from Klaver et al., (2012), Keller et al., (2013) and Houben et al., (2014). The results
720 previously explained have shown that this method is robust enough to be applied to a natural
721 and complex material such as shale, and can be applied to numerical and artificial materials
722 as well as natural materials.

723 The comparison of the phase area fractions for various sizes of domain can also be compared
724 with XRD results, to quantify if the mesoscale can be representative of the macroscale. XRD
725 analyses are performed on a 100 mg powder, but the powder comes from a centimeter-size
726 sample in order to represent as well as possible the macroscale. Area fractions from SEM
727 images represent the mesoscale. However, to compare weight and volume data from XRD
728 with area fractions from SEM images, an accurate 3D characterization of the microstructural
729 anisotropy is ideally required. 3D microstructural data are poorly documented at the
730 mesoscopic scale for the Bowland Shale, despite recent work at the microscale (Ma et al.,
731 2016). This comparison was not possible in this case.

732 **6.4. Using the geometrical microstructure models**

733 The geometrical models generated in this study represent the mesoscale of two laminae of a
734 sample of the Lower Bowland Shale, in which inclusions of quartz, feldspars, carbonates,
735 micas, heavy minerals, organic matter and fractures are embedded in a clay matrix and quartz
736 cement. The cement, clays, organics and microfractures define a porous network which
737 provides potential pathways for gas transport following fracturing stimulation. Frey et al.,
738 (2012) and van den Eijnden et al., (2016, 2017) use such microstructures to model the micro
739 and macroscale hydro-mechanical behavior of shale in a finite element squared (FE²) model,
740 attributing mechanical and petrophysical properties (e.g. stiffness, hydraulic conductivity) to
741 each phase. With microstructural parameters, Houben et al. (2014), Lan et al., (2010) and
742 Guo et al., (2013) also modeled the microstructure of shales and brittle rocks, by quantitative
743 approaches or by using a distinct element code to generate a deformable microstructure.

744 The model of microstructure of the Bowland Shale presented here can potentially act as a
745 basis for similar hydraulic transfer modeling in the Bowland Shale at the mesoscopic scale. It
746 can be used to upscale the hydromechanical behavior from the microstructure scale to the
747 macroscale taking into account the anisotropy of the small components of the shale material.

748 Moreover, the geometry of the microstructural model includes two scales: the scale relative to
749 individual grains and the scale of grain aggregates. It can therefore be used with whole
750 representative images or divided in small subareas (smaller “REA”) to study the influence of
751 microstructural heterogeneities (aggregates, individual grains in the cement, grains of
752 feldspars and quartz stacked together...) on the hydromechanical behavior of the material.

753 **6.5. Potential of image mosaics and representative microstructure models** 754 **for upscaling**

755 Microstructural data are collected at an infinitesimally smaller scale than the size of the
756 Bowland Basin, qualitatively estimated at the kilometer scale (Andrews, 2013). It is mainly

757 due to the fact that microstructural features are currently visible with high resolution
758 techniques such as microscopy and tomography, which do not allow the observation of larger
759 samples than the centimeter-size. However, numerical models for hydraulic fracturing require
760 representative microstructural data from the pore scale to hundreds of meters, which makes
761 upscaling a real challenge. Averages of microstructural parameters at a defined scale can be
762 calculated on representative areas of smaller scales and implemented into numerical
763 calculations at a larger scale (e.g. Wen and Jaime Gomez-Hernandez, 1996; Zhang et al.,
764 2012; Peng et al, 2015). Different methods such as homogenization or multiscale asymptotics
765 can be applied during the approach of upscaling (Khalili et al., 2012; Kazemi et al., 2012;
766 Davit et al., 2013).The geometrical models of the microstructure determined from
767 representative experimental areas have the same area fraction and the same distributions for
768 grain elongation ratio and orientation. However, they do not have the same grain structure
769 (layout). The dispersion method could therefore provide the quantification of the variability
770 of grain layout in those REAs. A standard deviation of particle layout could be used to define
771 a standard of deviation of the upscaled data resulted from these models. A few example
772 geometrical models are given in Appendix.

773 **7. Conclusions**

774 The microstructure of a laminated sample of Lower Bowland Shale from the Preese-Hall 1
775 borehole was mapped using a mosaic of 2D-SEM images at the mesoscopic scale. The
776 quantitative characterization of the sample's microstructure demonstrates a high vertical
777 variability of mineral content, grain properties and grain shape anisotropy at the millimeter
778 scale from a gradual organic-matter poor lamina to an organic-rich lamina. Quartz, feldspars
779 and carbonate aggregates act to increase the spatial variability inside each lamina. The multi-
780 scale 2D mapping of phase area fraction highlights multi-scale interfaces of area fractions,

781 which are potentially of importance to the understanding of fracture initiation and
782 propagation. The Lower Bowland Shale sample chosen here might have potential for multiple
783 direction fracture propagation due to the high variability of microstructure heterogeneities
784 and interfaces (subject to the anisotropy of the ambient stress field), in addition to a complex
785 natural fracture network that might be re-stimulated.

786 The counting-box, dispersion, covariance and 2D mapping approaches were compared to
787 estimate the representativeness of the microstructure of each lamina, based on the phase area
788 fraction parameter. 2D phase mapping supports the use of the dispersion approach to
789 calculate the REA in such heterogeneous rock on a large surface. Nevertheless the covariance
790 allows the quantification of a minimum homogeneous surface relative to the size of particles.
791 The models of the geometry of the microstructure were built with parameters calculated from
792 thousands of grains on a surface of $1.280 \times 1.682 \text{ mm}^2$, which is 3 to 11 times larger than the
793 maximum size of microstructural heterogeneities according to the four methods. This
794 microstructure model has been shown to be as representative as is possible from a sample of
795 this scale, and could be used as a starting point in the improvement of our understanding of
796 fluid flow through the porous phase in the Bowland Shale at the mesoscopic scale.

797

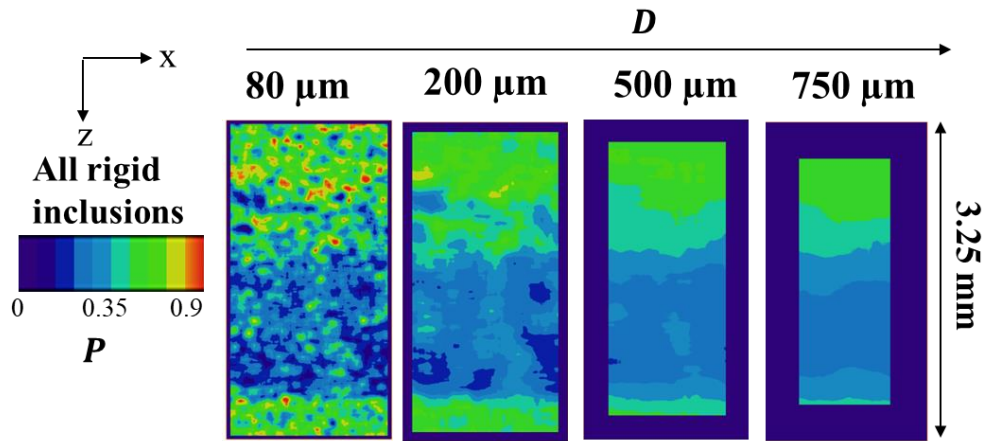
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803 Data Statement: representative research data is presented in the images and graphs in this
804 manuscript. Other datasets generated and/or analyzed during this study are not publicly

805 available due to their large size but are freely available from the corresponding author on
806 reasonable request.

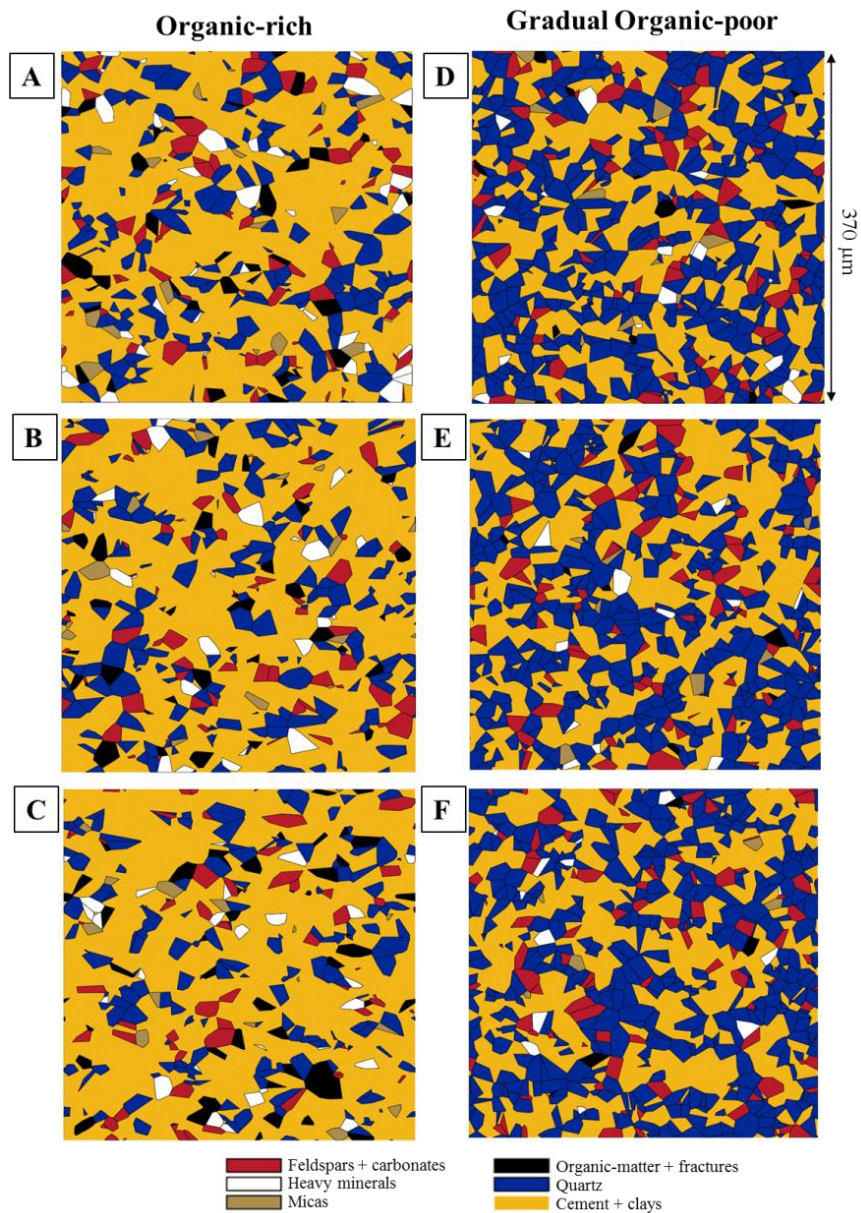
807

808 **Appendices:**



809

810 **Appendix 1 : Quantitative maps of the local area fraction for all rigid inclusions in the cement**
811 **(quartz+carbonates+feldspars+micas+heavy minerals+organics+fractures), as a function of the size of the**
812 **calculation domain D .**



813

814 **Appendix 2 : Example models of the geometry of the microstructure in the Lower Bowland Shale: A,B,C are random**
 815 **microstructures for the organic-rich lamina and D,E,F for the organic-poor lamina.**

816

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