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Philippe Cosenza, Anne-Laure Fauchille, Dimitri Prêt, Stephen Hedan, Albert
Giraud

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**Statistical Representative Elementary Area of Shale Inferred by
Micromechanics**

Intended for publication in International Journal of Engineering Science

23 **Abstract**

24 This paper provides new estimates for the Representative Elementary Area (REA) sizes of the
25 following two shales that are actively being studied in the framework of deep disposal of
26 radioactive waste: Callovo-Oxfordian (COx) claystone from the Meuse/Haute-Marne
27 underground research laboratory (Eastern France) and Toarcian argillite from the experimental
28 station of Tournemire (Southern France). The REA sizes, named L_{REA} , were obtained from two
29 mineral maps following the classic “counting box” (CB) method and a statistical approach that
30 introduces the concept of a “statistical” REA. Following this approach, a “statistical” REA is
31 related not only to the microstructure and the properties of each of the components but, above
32 all, to a given precision in the estimation of the effective property depending on the number of
33 realizations that one is ready to generate. The probabilistic concept of realization is here, from
34 a practical viewpoint, a subdomain of a mineral map in which the apparent morphological or
35 mechanical properties have to be calculated. In this study, the apparent elastic moduli of the
36 subdomain have been estimated using two micromechanical models. The first
37 micromechanical model consisted of an inclusion-based model for which spherical nonclay
38 grain is embedded in a clay matrix in which the values of its transverse isotropic stiffness tensor
39 have been taken from literature. The second micromechanical model was an isotropic
40 inclusion-based model for which a spherical nonclay grain is embedded in a clay matrix; the
41 elastic moduli values have been inverted by a Monte-Carlo approach from the engineering
42 moduli of both shales under study. Our calculations have shown the following results: (i) the
43 statistical morphological L_{REA} considering the surface clay fraction are of the same order of
44 magnitude as those measured in other shales and those obtained by the simple CB method,
45 with relative error values between 5 and 10%, (ii) the mechanical L_{REA} values associated with
46 the bulk modulus and shear modulus are significantly greater than that of the morphological
47 L_{REA} , and (iii) the mechanical L_{REA} estimates of the shear modulus are greater than that of the

48 bulk modulus. Moreover, our study highlights that these qualitative results do not depend on
49 the chosen micromechanical models and, thus, would be independent of the underlying
50 anisotropic nature of shale.

51 **Keywords:** microstructure, shales, clay-rocks, anisotropy, clay, elastic properties.

52 **Short title:** Mechanical REA of shales inferred by micromechanics.

53

54 **Highlights**

- 55 • New estimates for the REA sizes of COx claystone and Tournemire argillite.
- 56 • Mechanical REA size is greater than morphological REA size.
- 57 • REA sizes of Tournemire argillite are greater than those of the COx map.
- 58 • REA sizes of shear modulus are greater than those of the bulk modulus.
- 59 • These results do not depend on the chosen micromechanical models.

60

61 1. Introduction

62 Clay rocks, often called shales, are being studied as potential host rocks for high-level
63 radioactive waste repositories in several industrial countries (e.g., ANDRA, 2005; Pusch, 2006)
64 and form the main geological caps of many hydrocarbon reservoirs (e.g., Aplin and Larter,
65 2005). Organic-rich clay rocks are considered to be the key petroleum source rocks for the
66 coming decades (e.g., Klaver et al., 2015). These geological formations are highly
67 heterogeneous at multiple scales, but a major proportion of the literature agrees that the
68 following relevant microstructural levels must be considered (e.g., Bennett et al., 1991;
69 Jakobsen et al., 2003; Ulm et al., 2005; Ortega et al. 2007; 2009; Bobko and Ulm, 2008; Loucks
70 et al., 2012; Cariou et al., 2013; Keller, 2015) (Figure 1):

- 71 • Level 0 is the scale of elementary clay layers.
- 72 • Level 1 is the scale for which the elementary clay layers are packed together to
73 form clay particles.
- 74 • Level 2 is the submicrometer scale, often called the “microscopic” scale of
75 porous clay composites constituted in a mixture of clay particles or aggregates.
- 76 • Level 3, often called the “mesoscopic” scale in geosciences, is the scale where
77 the characteristic size is in the submillimeter range. At this scale, the rock is
78 considered to be a porous clay matrix mixed with a population of nonclayey
79 grains (mainly quartz, carbonates and pyrite). This is the scale of the mineral
80 maps used in this work.
- 81 • Level 4 is a lamina type that is associated with an alternation of clay-rich layers
82 and non–clayey materials (mainly quartz and carbonates).

83 With regards to the complexity of this hierarchical organization, the prediction of the
84 engineering properties of shale from its microstructure is clearly a challenging task. In the
85 following work, we will focus on the mesoscopic scale for which numerous theoretical studies

86 have been devoted to the calculations of the engineering or effective properties (e.g., Jakobsen
87 et al., 2003; Levin and Markov, 2005; Ulm et al., 2005; Giraud et al., 2007a, 2007b; Ortega et
88 al. 2007; 2009; Cariou et al., 2013; Cosenza et al., 2015a,b). This mesoscopic scale is often
89 considered the key scale for modeling the mechanical and transport behaviors of clay rocks
90 involved in high-level radioactive waste repositories (Abou-Chakra Guéry et al., 2010; Robinet
91 et al., 2007; 2012; Cariou et al., 2013). These effective properties, which are inferred by
92 numerical upscaling techniques or by homogenization theories, are used as inputs for numerical
93 codes, often based on the finite elements method, for engineering purpose.

94 All the upscaling or homogenization approaches used to estimate these effective
95 properties are based on the paramount concept of the Representative Elementary Volume
96 (REV), which is required to “separate” the following two space scales: the scale of
97 heterogeneity, i.e., nonclayey grains (or rigid inclusions) in our case, and the scale of
98 engineering for which clay rock is viewed as a continuum of homogeneous medium. Thus, the
99 REV is usually considered as a volume of heterogeneous material that is sufficiently large to
100 be statistically representative of the rock, i.e., to include a relevant sampling of all the
101 microstructural heterogeneities existing in the rock at the scale of interest (here, the mesoscopic
102 scale) (e.g., Kanit et al., 2003; Zhang et al., 2017). At the same time, the REV has to be small
103 enough, compared to the scale of the macroscopic geotechnical systems of study, “so that it
104 may be considered as infinitesimal in the mathematical treatment” (Biot, 1941).

105 To our knowledge, the approaches used to estimate the size of a REV, named hereafter
106 L_{REV} , can be divided for the sake of simplicity into two groups that introduce the probabilistic
107 concept of realization; here, a realization is any representation of the microstructure considered
108 with a given size and a given fraction of heterogeneities. The first approach or group of
109 approaches aims at using statistical parameters, e.g., mean, standard deviation or covariance, of
110 a given geometrical or physical property (e.g., porosity, fraction of a given mineralogical phase,

111 mechanical property, etc.) calculated over a significantly large set of realizations to
112 quantitatively describe the heterogeneous structure of the material of interest (Torquato and
113 Stell, 1982; Zhang et al., 2000; Kameda et al., 2006; Houben et al. 2014; Klaver et al., 2015;
114 Fauchille et al., 2018). Following these approaches, REV is defined as the elementary volume
115 below which the property of interest varies significantly across the scale. Among these
116 approaches, the “counting box” method and the covariance or the two-point probability function
117 are likely the most popular and have been recently applied to 2D images acquired on shale
118 samples (i.e., Pasidonia shale, Germany, Klaver et al., 2015; Callovo-Oxfordian argillites,
119 France, Song et al., 2015; Opalinus clay, Switzerland, Keller et al., 2013; Houben et al. 2014;
120 Bakken shale, United States of America, Liu and Ostadhassan, 2017; and Bowland shale,
121 United Kingdom, Fauchille et al., 2018). It should be noted that most of these works have been
122 performed on 2D images and, thus, provided estimations of the Elementary Representative Area
123 (REA) size, hereafter named L_{REA} .

124 This first group of approaches is often referred to as “deterministic” approaches
125 (Rolland du Roscoat, et al., 2007; Khdir et al., 2012), in contrast with a second group of
126 approaches that explicitly introduce the concept of the “statistical” REV. Following Kanit et al.
127 (2003) and Jeulin et al. (2004), a “statistical” REV is related not only to the microstructure and
128 the physical properties of each components but, above all, to a given precision in the estimation
129 of the effective property depending on the number of realizations “that one is ready to generate”
130 (Jeulin et al., 2004). Consequently, in comparison with the aforementioned approaches, the
131 “statistical” L_{REV} depends explicitly on an additional parameter, i.e., the precision desired for
132 the estimate of the effective property and reached for a given number of realizations.
133 Consequently, the estimate of the “statistical” L_{REV} cannot be unique without any associated
134 precision and number of realizations. This statistical approach has been recently applied to

135 Opalinus clay samples taken from the Mont Terri rock laboratory in Switzerland (Houben et
136 al., 2014; Keller, 2015; 2016a,b).

137 In summary, this brief review shows that a large variety of methods used to infer L_{REV}
138 or L_{REA} estimates with regard to shale exist, and these estimates concern mainly the
139 petrographical or mineralogical parameters of the porosity and mineral phases. Most of them
140 have been obtained by deterministic approaches, e.g., the counting box method and dispersion
141 method based on the standard deviation calculations. Moreover, to our knowledge, estimates of
142 the mechanical L_{REV}/L_{REA} of shales are very scarce; one recent and single work was performed
143 on Opalinus clay by Keller (2016a,b), and it focused on the dynamic elastic moduli.

144 The objective of this paper is thus twofold. First, we would like to provide new estimates
145 of L_{REA} obtained from two mineral maps (Jorand, 2006; Fauchille, 2015) acquired from two
146 shales extensively studied in the framework of the deep disposal of radioactive waste, as
147 follows: Callovo-Oxfordian (COx) claystone from the Meuse/Haute-Marne underground
148 research laboratory (Eastern France) and Toarcian argillite from the experimental station of
149 Tournemire (Southern France). These estimates have been obtained by the classical counting
150 box method and the statistical approach promoted by Kanit et al. (2003; 2006) and Jeulin et al.
151 (2004). Second, we would like to quantify the statistical mechanical L_{REA} of COx claystone and
152 Tournemire argillites by focusing on static elastic parameters for further engineering purposes.
153 These mechanical L_{REA} are inferred by using micromechanical approaches, one of which
154 implicitly accounts for the transverse isotropic nature of shale.

155 **2. Mineral maps**

156 The L_{REA} estimates calculated in this study have been obtained from two mineral maps
157 acquired following the methodologies described below.

158 The first mineral map, hereafter called the COx map, has been made from a sample
159 taken from the Callovo-Oxfordian (COx) claystone, which is under extensive study at the

160 Meuse/Haute-Marne Underground Research Laboratory (MHM-URL) (Eastern France). The
161 thickness of this formation is 130 m and its age is 150-160 My; the formation is located 420-
162 550 m below the surface, in the eastern part of the Paris Basin (Andra, 2005). The Callovo-
163 Oxfordian formation contains mainly 25 to 65 wt.% clay minerals, with 20-42 wt.% carbonates
164 (calcite, dolomite, ankerite) and 15-31 wt.% quartz and feldspars (Andra, 2005). Considering
165 nonclay minerals, feldspars contents are negligible regardless to quartz. Among carbonates,
166 siderite and dolomite are negligible regardless to calcite. Thus, quartz and calcite will be the
167 only nonclay minerals considered in the following.

168 This mineral map was prepared from a drill-core, denoted as EST05-709 (-492.2 m) and
169 extracted from the Andra EST205 borehole (Jorand, 2006; Conil et al., 2018). It was obtained
170 at a micrometer spatial resolution from an advanced image processing of a chemical elements
171 map that was acquired through the use of a Cameca SX100 electron probe microanalyzer (Prêt,
172 2003). This electron microanalyzer provides quantitative concentration maps of 14 chemical
173 elements (Al, Na, K, Ca, Si, Mg, Ti, Fe, S, Ba, Zr, P, Zn, Sr) on a 3 x 0.5 mm² area with a
174 spatial resolution of 2 µm/pixel. The image processing of these maps is based on mineral
175 thresholding methods that accommodate mixtures and solid solutions. For details, the reader is
176 referred to Prêt (2003) and Prêt *et al.* (2010a,b). In our case, this methodology allows for the
177 spatial location of 16 different minerals, including the following three different clay minerals:
178 illite-smectite mixed layers, kaolinite and chlorite.

179 The geometrical and mineralogical features of the COx map are given in Table 1. The
180 surface fractions of the clay matrix, calcite and quartz are 50.4%, 25.0% and 13.8%,
181 respectively. Note that the largest side (1563 pixels, 3072 µm) and the smallest side (250 pixels,
182 500 µm) are perpendicular and parallel to the bedding, respectively.

183 The second mineral map, hereafter called the Toar map, has been extracted from a large
184 mosaic acquired from a nonimpregnated and dried Tournemire clay rock sample (Fauchille et

185 al., 2014; Fauchille, 2015). The Tournemire clay rock sample studied has been sampled in the
186 horizontal and cylindrical borehole FD90 in the 1996 East gallery of the Tournemire
187 Underground Research Laboratory (URL) of the French Institute for Radioprotection and
188 Nuclear Safety (IRSN). The sample was located at a depth between 4.20 to 4.40 meters far from
189 the gallery wall, outside the so-called Excavation Damaged Zone. The Tournemire URL is
190 located in a Mesozoic basin on the southern border of the Massif Central (Aveyron, France), in
191 the subhorizontal consolidated argillaceous Toarcian formation (200 meters thick) and marly
192 layers of the Domerian age (50 meters thick). The sample comes from the upper Toarcian
193 formation, whose mineralogical composition shows that clay minerals represent nearly 25-70
194 wt% of the rock with illite (10-40 wt%) and illite/smectite mixed-layer minerals (5-25 wt%),
195 kaolinite (10-35 wt%) and chlorite (1-5 wt%). The Tournemire clay rock also contains 10-40
196 wt% of carbonates, 10-30 wt% of quartz, 2-7 wt% of sulfides and less than 2 wt% of feldspars
197 (Cabrera et al., 2001). Similarly to COx map, quartz and calcite are the dominant nonclay
198 minerals and will be the only nonclay minerals to be considered.

199 The mosaic of interest (7.1 x 5.2 mm², 11302 x 8355 pixels) has been built from one
200 hundred and fifty three back-scattered electron images (spatial resolution of 0.625 μm.pixel⁻¹)
201 acquired by Scanning Electron Microscopy (SEM, JEOL JSM 56000LV with an acceleration
202 voltage of 15 kV, a probe current of 5 nA, a working distance of 16.3 mm, a magnification of
203 x200, and a dwell time of 128 μs per pixel). On the mosaic of images, the clay-matrix,
204 carbonates, quartz, sulfides and macropores were differentiated by the MicroPhaseMap©
205 software developed at the IC2MP laboratory in Poitiers, allowing the boundary between the
206 clay matrix and nonclay grains to be determined (Prêt et al., 2010a,b). The mineral map used
207 in this study concerns an extraction (4000x4000pixels) of the most homogeneous part of the
208 mosaic in order to respect at best the statistical homogeneity assumed in all the further *L_{REA}*
209 calculations. The geometrical and mineralogical features of this map are also given in Table 1.

210 Extractions of both maps are given in Figures 2 and 3. Note that both maps were
211 prepared from a polished thin section in a plane perpendicular to the stratigraphic plane. The x-
212 direction indicated in Figures 2 and 3 is parallel to the bedding planes, whereas the z-direction
213 is perpendicular to the bedding.

214 The results of these image analyses are given in numerical files in which the location
215 and mineral code of each pixel of the mineral maps are indicated. These numerical files are the
216 input files for the L_{REA} estimate calculations that are presented in the next section.

217 **3. Methodologies used to infer the REA size**

218 *3.1 Counting box method*

219 The counting box method, which is likely the most popular to infer L_{REA} , starts from a
220 given subdomain or box in the digitalized image. Then, the mean of a surface property (surface
221 mineral contents, surface porosity, physical property, etc.) is calculated within increasing
222 subdomains, often called boxes, until the actual image size is reached (Houben et al., 2014;
223 Klaver et al., 2015). The characteristic size L_{REA} is considered to be reached when the mean of
224 the considered property does not evolve significantly with the increasing size of boxes. This
225 procedure can be repeated for several starting subdomains to make sure that the inferred L_{REA}
226 is statistically representative of the whole image. This first method will be named hereafter the
227 classical counting box (CB) method.

228 Regarding the use of square domains in the CB method, the COx and the Toar maps
229 have been divided into six and four nonoverlapping square areas, respectively, following the
230 partitioning given in Figure 4. These nonoverlapping areas, named A_i ($i=1,\dots,6$ for COx map;
231 $i=1,\dots,4$ for Toar map), are associated with the starting of the subdomains, which are defined
232 and discriminated by the coordinates of their center C_i ($i=1,\dots,6$ for COx map; $i=1,\dots,4$ for Toar
233 map) (Figure 4). Note that the origin, i.e., $x=0$, $z=0$ of the system of coordinates is located in
234 the top left corner of both maps (Figures 2, 3 and 4).

235 The particular partitioning displayed in Figure 4 is due to two reasons. First, the shape
236 of the COx map is elongated following the z-axis and the center of the initial box could not be
237 located only at the center of this map to investigate the whole map. Second, it was interesting
238 from a statistical viewpoint to compare the L_{REA} estimates calculated at different areas with a
239 comparable surface and, thus, to check the statistical homogeneity of each map. Note that the
240 number of pixels of each area is sufficient to apply the deterministic CB approach.

241 *3.2 “Statistical” Elementary Representative Area (REA)*

242 Kanit et al. (2003; 2006) and Jeulin et al. (2004) extended the classic definition of REV
243 by explicitly integrating the concept of “desired precision” into the calculated effective
244 property. Following their approach, the effective physical properties of heterogeneous materials
245 can be determined not only by one single numerical simulation on a large volume (greater than
246 the REV) of the microstructure but also as the mean value of apparent properties calculated on
247 volumes smaller than the REV, provided than a sufficient number of realizations of the
248 microstructure are considered (e.g., Rolland du Roscoat et al., 2007). Therefore, the “statistical”
249 REV introduced in this manner is not unique; it is defined as a function of the physical
250 properties of each component, the microstructure, the desired precision and the number of
251 realizations. This methodology was used to determine the “statistical” REVs of mechanical and
252 thermal properties of specific random microstructures (Kanit et al., 2003; Pelissou et al., 2009)
253 and of real microstructures of clay rock (Houben et al., 2014; Keller, 2015; 2016a,b). In our
254 study, we will focus on elastic moduli of shale considered as a transverse isotropic rock.

255 The determination of “statistical” REVs is obtained following a two-step procedure. In
256 the first step, the paramount concept of this approach, the integral range A_3 (or A_2 by considering
257 a 2D image) has to be evaluated.

258 *3.2.1 Determination of integral range*

259 Considering a 2D microstructure, the integral range A_2 , which depends on the studied
 260 property, is directly related to the scatter (the dispersion) in apparent properties calculated from
 261 several realizations n of the microstructure of fixed size S . The integral range gives information
 262 on the area size of the microstructure, for which the property measured in this area has a
 263 convenient statistical representativity (e.g., Matheron, 1971; Lantuéjoul, 1991).

264 For an ergodic, stationary random function $Z(x)$, and for a large specimen ($S \gg A_2$), the
 265 integral range A_2 can be obtained by the following expression (appendix; see also Lantuéjoul,
 266 1991; Kanit et al. 2003; 2006 and Jeulin et al., 2004) as follows:

$$267 \quad D_Z^2(S) = D_Z^2 \frac{A_2}{S} \quad (1)$$

268 where $D_Z^2(S)$ is the variance of the mean value \bar{Z} over the surface S and D_Z^2 is the point
 269 variance of $Z(x)$. Equation (1) is valid for additive properties (e.g., surface fraction or mass
 270 density) and, thus, can be rewritten as follows when the surface clay fraction, namely, φ is
 271 considered:

$$272 \quad D_\varphi^2(S) = \bar{\varphi}(1 - \bar{\varphi}) \frac{A_2}{S} \quad (2)$$

273 where the variance D_Z^2 is $\bar{\varphi}(1 - \bar{\varphi})$. As the composition of a physical property $Z(x)$
 274 (e.g., elastic moduli in our case) in the change of scale is not additive, the latter relation (2) is
 275 no more valid, and Kanit et al. (2003) proposed the following modified version:

$$276 \quad D_Z^2(S) = D_Z^2 \left(\frac{A_2}{S} \right)^\alpha \quad (3)$$

277 where α is an exponent that has to be determined. In the case of a two-phase material
 278 with the physical property Z_1 for phase 1 and Z_2 for phase 2, the point variance of the random
 279 variable Z is as follows (Kanit et al. 2003; and Jeulin et al., 2004):

$$280 \quad D_Z^2 = P(1 - P) (Z_1 - Z_2)^2 \quad (4)$$

281 where P is the volume fraction of phase 1 or phase 2. In practice, phase 1 and phase 2
 282 are the clay matrix and nonclay inclusions, respectively. In this study, this means that properties

283 Z_1 and Z_2 are the elastic modulus, either the bulk modulus or shear modulus, of the nonclay
 284 inclusions and clay matrix, respectively.

285 As illustrated further, in both cases, i.e., considering the clay fraction φ (equation 2) and
 286 a chosen physical property Z (equation 3), the integral range A_2 is obtained by fitting from the
 287 graph of the function $D_\varphi^2(S)$ or $D_Z^2(S)$. In our case, let us recall that Z is an elastic modulus,
 288 either the bulk modulus or shear modulus, the variance $D_Z^2(S)$ of which is calculated on the
 289 partitions of the mineral map in subdomains of a given area S . The value of Z for a given
 290 subdomain of area S is obtained from the micromechanical approaches presented in the
 291 following section 3.2.2.

292 3.2.1 Determination of the REA size

293 In a second step, the determination of the integral range allows the calculation of the
 294 precision desired to estimate the size L_{REA} . In the theory of samples, the absolute error ε_{abs} and
 295 the relative error ε_{rel} of the mean effective value \bar{Z} obtained with n independent realizations of
 296 area S are deduced from the interval of confidence as follows (e.g., Jeulin et al., 2004):

$$297 \quad \varepsilon_{abs} = \frac{2 D_Z(S)}{\sqrt{n}} \quad (5)$$

$$298 \quad \varepsilon_{rel} = \frac{\varepsilon_{abs}}{\bar{Z}} = \frac{2 D_Z(S)}{\bar{Z}\sqrt{n}} \quad (6)$$

299 Thus, the REA area, S_{REA} , can now be defined as the area for which n realization ($n=1$
 300 for instance) is required to estimate the mean effective property \bar{Z} with a given relative error
 301 ($\varepsilon_{rel}=5\%$ for instance) provided that the function $D_Z(S)$ is known.

302 For instance, if Z is the surface clay fraction named φ , the parameter S_{REA} , estimated
 303 with a given relative error ε_{rel} and a given number of realization n , can be obtained by combining
 304 equations (6) and (2), as follows, provided that the mean effective property $\bar{\varphi}$ is known:

$$305 \quad S_{REA} = L_{REA}^2 = \frac{4(1 - \bar{\varphi})A_2}{n \varepsilon_{rel}^2 \bar{\varphi}} \quad (7)$$

306 In the same way, if Z is now a physical property, the corresponding parameter S_{REA} ,
 307 estimated with a given relative error ε_{rel} and a given number of realization n , can be deduced
 308 by combining equations (3), (4), (5) and (6), as follows, provided that the mean effective
 309 property \bar{Z} is known:

$$310 \quad S_{REA} = L_{REA}^2 = \left[\frac{4P(1-P)(Z_1 - Z_2)^2}{n \varepsilon_{rel}^2 \bar{Z}^2} \right]^{1/\alpha} A_2 \quad (8)$$

311 At this stage, two remarks have to be formulated. First, the integral range A_2 in both
 312 equations (7) and (8) are not *a priori* similar. The integral range A_2 depends on the property
 313 under study, i.e., φ or Z , and has to be evaluated separately. Second, this approach assumes that
 314 the mean effective properties $\bar{\varphi}$ and \bar{Z} are known *a priori*. In practice, the latter are calculated
 315 on macroscopic areas supposed to be much greater than the area of the REA.

316 In summary, a “statistical” REV is therefore determined with the following algorithm
 317 (Kanit et al., 2003):

- 318 i) Consider different realizations of the microstructure, i.e., different partitions of
 319 the mineral map in subdomains of a given area S .
- 320 ii) Calculate the mean and the variance of the property of interest of the whole set
 321 of partitions under consideration; the graph of the function $D_{\varphi}^2(S)$ or $D_Z^2(S)$ is
 322 thus obtained.
- 323 iii) Estimate the integral range A_2 from the previous graph, equation (2) or equation
 324 (3) (in the later, the exponent α has to be evaluated as well).
- 325 iv) Define the precision ε_{rel} and the number of realization n to estimate the effective
 326 property of interest and calculate the L_{REA} with equation (7) or equation (8). The
 327 value of the effective property, $\bar{\varphi}$ or \bar{Z} in equation (7) or equation (8), is
 328 independently estimated.

329

330 3.3.2 Calculations of apparent elastic properties

331 As mentioned previously, the calculation of the integral range A_2 associated with a given
332 mechanical property requires the values of the average and the variance of the mechanical
333 property calculated over a set of subdomains of the map of interest (step ii). This means that
334 the mechanical property under study has to be calculated in each subdomain of size S of the
335 map partition. The apparent mechanical property associated with a given subdomain has been
336 estimated in this work by two micromechanical approaches.

337 *Anisotropic model*

338 Following the first micromechanical approach, hereafter called the anisotropic model,
339 the transverse isotropic nature of shale is implicitly taken into account. This anisotropic model
340 is itself achieved in two steps. In a first step, the apparent anisotropic stiffness tensor in the
341 subdomain of interest is calculated by an inclusion-based model, i.e., a nonclay spherical grain
342 embedded in a clay transverse isotropic matrix following a differential effective medium
343 (DEM) scheme (e.g., Cosenza et al., 2015b). The latter scheme is widely and successfully used
344 in rock physics to model the elastic wave velocities of numerous rocks (e.g., Hornby et al.,
345 1994; Markov et al., 2005).

346 The DEM scheme consists of iteratively adding spherical inclusions with diluted
347 concentration into the current effective medium determined in the previous step. At the initial
348 stage, a material with elastic moduli C_M is considered as the initial host material. A dilute
349 concentration of spherical inclusions is then added into this initial host; the effective tensor of
350 this new composite can be calculated without considering the interactions between the
351 inclusions. The construction process continues as follows: (a) at each stage, the embedded
352 spherical inclusions are scattered in diluted concentration and (b) the required volume fraction
353 of inclusions is satisfied. At a given step, indexed i , the small increment of stiffness due to the
354 addition of a small fraction of inclusions, df_I is given by the following:

$$d\mathbf{C}_i^{ap} = \frac{df_I}{1-f_I} (\mathbf{C}_I - \mathbf{C}_i^{ap}) : [1 + \mathbf{P}_i : (\mathbf{C}_I - \mathbf{C}_i^{ap})] \quad (9)$$

This increment $d\mathbf{C}_i^{ap}$ must be added to the apparent tensor \mathbf{C}_i^{ap} obtained in the previous step. Note that the bold capital letters refer here to fourth-order tensors. In our case, the stiffness tensors \mathbf{C}_I and \mathbf{C}_M are the stiffness tensor of the isotropic nonclay sphere and the stiffness tensor of the transverse isotropic clay matrix, respectively. The Hill's tensor \mathbf{P}_i is a function of \mathbf{C}_i^{ap} and the Eshelby's tensor, which depends on the chosen inclusion-based model, as follows:

$$\mathbf{P}_i = \mathbf{S}_i : (\mathbf{C}_i^{ap})^{-1} \quad (10)$$

The expressions of components of the Hill's tensor for a spherical inclusion in a transversely isotropic medium have been taken from Withers (1989) and Giraud et al. (2007a,b).

In a second step, the apparent tensor \mathbf{C}^{ap} obtained by DEM is isotropized following the classical decomposition, as follows:

$$\mathbf{C}^{ap, is} = 3K^{is}\mathbf{J} + 2\mu^{is}\mathbf{K} \quad (11)$$

with

$$K^{is} = \frac{C_J^{ap, is}}{3} = \frac{J::\mathbf{C}^{ap}}{3} ; \mu^{is} = \frac{C_K^{ap, is}}{10} = \frac{K::\mathbf{C}^{ap}}{10} \quad (12)$$

$$\mathbf{C}_J^{ap, is} = \frac{1}{3}\mathbf{C}_{ii jj}^{ap, is} ; \mathbf{C}_K^{ap, is} = \mathbf{C}_{ij ij}^{ap} - \frac{1}{3}\mathbf{C}_{ii jj}^{ap, is} \quad (\text{summation on repeated indices } i, j) \quad (13)$$

and four-rank tensors \mathbf{J} and \mathbf{K} obey the following algebraic properties:

$$\mathbf{J} : \mathbf{J} = \mathbf{J} ; \mathbf{K} : \mathbf{K} = \mathbf{K} ; \mathbf{K} : \mathbf{J} = \mathbf{J} : \mathbf{K} = \mathbf{0} \quad (14)$$

With

$$J_{ijkl} = \frac{1}{3}\delta_{ij}\delta_{kl} ; I_{ijkl} = \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) ; K_{ijkl} = I_{ijkl} - J_{ijkl} \quad (15)$$

This "isotropization" process of apparent stiffness tensor \mathbf{C}^{ap} is due to two reasons. The first reason is theoretical. The original methodology described in Kanit et al. (2003; 2006) and Jeulin et al. (2004) involves the isotropic physical properties in equations (3), (4) and (8). We do not know to what extent this methodology can be generalized to anisotropic media, i.e., to

378 mixtures of anisotropic constituents. In other words, this generalization to anisotropic media
379 has to be proven and its demonstration is beyond the scope of this work. Moreover, it should
380 be noted that despite this “isotropization” process, the calculation of the apparent stiffness
381 tensor of shale is impacted by that of the anisotropic stiffness tensor of the clay matrix and,
382 thus, implicitly depends on the anisotropic nature of rock. The second reason is methodological.
383 This “isotropization” allows the following results to be directly compared to those obtained by
384 a pure isotropic model presented further, since both models provide apparent moduli with the
385 same physical meaning.

386 Equations (11) to (15) allow the extraction of the apparent isotropized bulk modulus,
387 K^{is} and the apparent isotropized shear modulus μ^{is} , which are used to infer the statistical
388 mechanical REA sizes (i.e., using equation (8)). However, the calculations of K^{is} and μ^{is} and,
389 thus, the \mathbf{C}^{ap} components require input data, which are the components of the stiffness tensors
390 \mathbf{C}_I and \mathbf{C}_M .

391 The components of stiffness tensor \mathbf{C}_I correspond to the elastic moduli of nonclay grains
392 (quartz and calcite) whose values have been taken from Mavko et al. (2009). In our calculations,
393 the isotropic elastic moduli of the nonclay phase, i.e., the bulk and shear modulus, are the
394 weighted averages of those of quartz and calcite, where the weights are the gravimetric contents
395 of each phase, as indicated in Table 2. In fact, the pure calcite and quartz phases are anisotropic
396 minerals (e.g., Mavko et al., 2009); their isotropized equivalent elastic properties considered
397 here correspond to random distributions in the orientation and the space of calcite and quartz
398 inclusions (e.g., Ulm et al., 2005; Giraud et al., 2007b).

399 The elastic moduli of the clay matrix are more difficult to estimate, because the clay
400 phase is also a heterogeneous composite composed of aggregates of clay layers, pores filled
401 with interstitial fluids and, likely, fine nonclay minerals (Figure 1). Consequently, in practice,
402 the elasticity of the clay phase is complex, which makes it difficult to quantitatively characterize

403 in an experimental viewpoint, and, thus, is poorly constrained in our context. To our knowledge,
404 the most relevant range of transverse-isotropic elastic properties of a clay matrix at the
405 mesoscopic scale has been obtained by Ulm et al. (2005), who used an experimental
406 micromechanics approach by combining nanoindentation measurements with the
407 microporoelasticity theory. These authors have proposed quantitative estimations of elastic
408 moduli, which resulted from the inversion of nanoindentation measurements obtained on three
409 different shale materials at spatial scales of 10^{-7} to 10^{-5} m, which correspond to that of our
410 mineral maps (Ulm et al., 2005; Cosenza et al., 2015b). In our calculations, only the lower
411 bound of the range proposed by Ulm et al., (2005) has been considered (Table 2). Indeed, the
412 values of transverse isotropic moduli considered in Table 2 provided mean values of apparent
413 elastic moduli that are the closest to the reference macroscopic elastic moduli measured in both
414 shales under study. Higher values of clay elastic moduli would have led to much higher values
415 of apparent moduli that are significantly higher than the experimental elastic moduli measured
416 on both shales.

417 *Isotropic model*

418 In the second micromechanical approach used in this work, hereafter called the isotropic
419 model, the elastic properties of the clay matrix, K_M and μ_M , were not given *a priori*. They have
420 been inverted following a Monte-Carlo procedure to overcome the paramount difficulty of
421 quantitatively characterizing the clay phase elasticity. This inversion procedure has considered
422 the following as input data: the weighted averages of quartz and calcite, the elastic properties
423 of both shales and the reference macroscopic elastic properties, which are all given in Table 2.
424 It was based on a simple micromechanical model, i.e., an inclusion-based model consisting of
425 an isotropic sphere embedded in an isotropic matrix. This isotropic inclusion-based model
426 corresponded again to a DEM approach, which is expressed by the following coupled
427 differential equations (e.g., Christensen, 2012):

$$428 \quad dK^{ap} = \frac{df_I}{1-f_I} \frac{K_I - K^{ap}}{1 + \frac{K_I - K^{ap}}{K^{ap} + 4/3\mu^{ap}}} \quad (16)$$

$$429 \quad d\mu^{ap} = \frac{df_I}{1-f_I} \frac{15(1-\nu^{ap})(\mu^{ap} - \mu_I)}{7 - 5\nu^{ap} + 2(4 - 5\nu^{ap}) \frac{\mu_I}{\mu^{ap}}} \quad (17)$$

430 with

$$431 \quad \nu^{ap} = \frac{3K^{ap} - 2\mu^{ap}}{2(3K^{ap} + \mu^{ap})} \quad (18)$$

432 The Monte-Carlo procedure consisted of 10000 random trials of clay matrix moduli
 433 ranging between 0 GPa and the reference macroscopic values given in the first column in Table
 434 2. The maximum relative difference between the reference macroscopic value and the
 435 calculated macroscopic value by our inverse procedure was less than 2% (Table 3). This
 436 difference was considered acceptable with regard to the high uncertainty associated with the
 437 quantification of clay matrix elastic moduli.

438 4. Results and Discussion

439 4.1 Counting box REA sizes versus Statistical morphological REA sizes

440 REA sizes inferred by the counting box method

441 Figures 5 and 6 display the evolutions of the clay fraction calculated on increasing box
 442 sizes L considering the different starting domains of the COx map and Toar map, respectively.
 443 Both figures confirm that the calculated clay fraction is a decreasing function of L and
 444 converges to a value, hereafter referred to as the parameter φ_∞ , which is very close to the mean
 445 clay fraction obtained on the whole map (Table 1).

446 Considering the COx map (Figure 5a.), the six curves associated with six starting
 447 domains converge to φ_∞ values in the range [48.6-52.4%], including the mean clay fraction
 448 $\bar{\varphi}_{COx}$ of 50.4% calculated on the whole map (Table 1). The difference between these asymptotic
 449 values, φ_∞ , and the mean clay fraction $\bar{\varphi}_{COx}$ is less than 4%. In the same way, in Figure 5b, the

450 four curves associated with the Toar map converge to φ_{∞} values in the range [68.5-71.4%],
451 which also include the mean clay fraction $\overline{\varphi}_{Toar}$ of 69.9% calculated for the whole map (Table
452 1). The difference between the φ_{∞} values and $\overline{\varphi}_{Toar}$ is less than 2.5%.

453 Below 100 μ m and 200 μ m for COx and Toar maps respectively, the curves obtained for
454 the different subdomains presents non correlated and high frequency evolutions. Such a
455 behavior is associated to the occurrence of a few grains with such a large size (Robinet, 2008;
456 Fauchille, 2015) and that large enough box size including several grains should be used to
457 estimate a meaningful REA (Gaboreau et al 2016). For larger box size than 100 μ m and 200 μ m
458 for COx and Toar maps respectively, the gap between the curves decreases progressively with
459 low frequency variations. Meaningful REA corresponding to mesoscopic scale (level 3 in
460 Figure 1) could be estimated with an improved accuracy when the box size increases. The
461 careful observation of both maps reveals that grain size is larger for Toar than for COx (compare
462 calcite grains in Figure 2 and carbonates grains in Figure 3), somehow explaining why a larger
463 box size is needed for Toar map to reach a REA as illustrated below (Figures 5a and 5b).

464 The REA size, i.e., L_{REA} , of both maps has been estimated in two steps (Table 4). In the
465 first step, the REA sizes have been calculated for each of the nonoverlapping areas of both maps
466 (6 areas for the COx map and 4 areas for the Toar map) and for two errors or threshold values,
467 i.e., $\varepsilon=0.1$ (10%) and $\varepsilon=0.05$ (5%). For each area, the L_{REA} parameter has been identified as
468 the lowest box size L for which the calculated mean clay fraction was significantly similar to
469 that of the whole map ($\overline{\varphi}_{COx}$ or $\overline{\varphi}_{Toar}$), with a maximum error of ε . In a second step, the
470 average overall L_{REA} estimates of all nonoverlapping areas have been calculated for each map.
471 The calculated mean L_{REA} for a given map has been considered as the L_{REA} of the latter.

472 Our results given in Table 4 indicate that the L_{REA} values are dispersed and decreasing
473 functions of the chosen error or threshold values ε . Indeed, in the first case ($\varepsilon=0.1$), the mean
474 values of the L_{REA} estimates of the COx map and Toar map are 173 μ m and 129 μ m, respectively

475 (Table 2). Whereas in the second case ($\varepsilon=0.05$), the mean values of the L_{REA} estimates of the
476 COx map and Toar map are much higher, i.e., 234 μm and 441 μm , respectively.

477 This scatter in the L_{REA} estimates can also be evidenced by a calculation of the standard
478 deviation of the L_{REA} estimates associated with nonoverlapping areas (Table 4). Considering the
479 COx map, the standard deviation values of the L_{REA} estimates increase from 108 μm ($\varepsilon=0.1$) up
480 to 143 μm ($\varepsilon=0.05$). The standard deviation values of the L_{REA} estimates of the Toar map
481 increase from 72 μm ($\varepsilon=0.1$) up to 267 μm ($\varepsilon=0.05$) (Table 4).

482 This scatter questions the statistical homogeneity of both maps and could be explained
483 by a small but significant evolution of the microstructure in the x direction and/or in the z
484 direction (i.e., with depth); this is shown in (a), where the φ_{∞} estimates of the COx map,
485 globally increase with depth, and by (b), where the φ_{∞} estimates of the Toar map, decrease in
486 the x direction (Table 4).

487 However, the order of magnitude of all these estimates is comparable to the results from
488 other shales obtained also by the CB method (Opalinus clay: 180-250 μm , Houben et al., 2014;
489 Posidonia shale and Whitby shale: 200 μm , Houben et al., 2016; Bowland shale: 380 μm , Ma
490 et al., 2016; Bakken shale: 176 μm , Liu and Ostadhassan, 2017).

491 *Statistical morphological REA sizes*

492 Figure 6 displays the mean values of the surface clay fraction $\bar{\varphi}$ and their corresponding
493 standard deviation values, all calculated on sets of map partitions in subdomains of size L
494 ranging from 6 to 140 μm (COx map) and from 6.25 to 250 μm (Toar map). Note that these
495 sets of map partitions do not correspond to the partitions A_i indicated in Figure 4, which have
496 only been used for the CB method. All estimates of the statistical REA sizes in the following
497 have been obtained by considering the whole maps.

498 Figure 6 suggests the following two comments. First, whatever the subdomain size, the
499 mean values of the surface clay fraction (50.5% for COx map and 69.9% for Toar map) are
500 very close to that of the whole map ($\bar{\varphi}_{COx}$ or $\bar{\varphi}_{Toar}$). For large subdomain sizes, these mean
501 values are almost equal to $\bar{\varphi}_{COx}$ or $\bar{\varphi}_{Toar}$. Second, the dispersion of the results characterized
502 by their standard deviation decreases with increasing the subdomain size and tends to a plateau
503 for L values approximately equal to 100 μm and 140 μm , considering the COx map and the
504 Toar map, respectively (Figure 6). Both trends are similar to those already observed for other
505 materials, considering other microstructural properties (porosity and specific surface area)
506 (Zhang et al., 2000; Kanit et al., 2003, 2006).

507 The decrease in dispersion with L for both maps evidenced in Figure 6 is also illustrated
508 in Figure 7, which displays the evolution of the pseudovariance $D_{\varphi}^2(S)/(\bar{\varphi}(1 - \bar{\varphi}))$ as a
509 function of the subdomains area S (i.e., L^2). In Figure 7, a $1/S$ fit for a large S by the least-square
510 method is also indicated to obtain values of the integral range A_2 (equation (2)) required to
511 calculate estimates of the REA size, L_{REA} , for both maps (equation (7)). The results of the fits
512 for both maps and the L_{REA} estimates considering one realization ($n=1$) and a range of error [5-
513 10%] are given in Table 5. This table shows two results.

514 First, the L_{REA} estimates in Table 5 are rather close to those obtained by the CB method
515 with similar values of errors, i.e., between 5 and 10% (Table 4). Indeed, in the case of the COx
516 map, the statistical REA size, L_{REA} , is in the range of [205-410 μm], whereas L_{REA} , estimated
517 by CB method is in the range of [50-438 μm] (Table 4). In the case of the Toar map, the
518 statistical L_{REA} estimates are in the range of [345-696 μm], whereas the L_{REA} estimates by the
519 CB method are in the range of [68-749 μm]. However, the comparison between these two types
520 of estimate is no more relevant for smaller errors ε since the L_{REA} estimate inferred by the
521 statistical method drastically increases as a function of $1/\varepsilon$ (see equation (10)).

522 Second, whatever the ε value, these results demonstrate that the REA size of the
523 Callovo-Oxfordian claystone is lower than that of the Tournemire argillite. The REA size
524 estimates of the COx map are in the range of [205-410 μm], whereas those of the Toar map are
525 in the range of [345-696 μm]. This hierarchy has also been observed in Table 4 following the
526 CB method applied with a ε value of 5%.

527 Figure 8 displays the values of the statistical morphological REA size, L_{REA} , as a
528 function of error ε (%) for one realization ($n=1$) for both mineral maps. The gray area in Figure
529 8 indicates the L_{REA} estimates for errors in the range of [0.1-10%]. Figure 8 shows that the L_{REA}
530 estimates of the COx map are always lower than those of the Toar map, regardless of the ε
531 values.

532 *4.2 Statistical morphological REA sizes versus Statistical mechanical REA sizes*

533 Figures 9a and 9b display the evolution of the mean values of the mechanical moduli
534 (bulk modulus and shear modulus) considering both the anisotropic and isotropic models as a
535 function of the size L of subdomains S . These figures confirm two trends previously observed
536 in Figure 6. First, the mean values \bar{K} and \bar{G} tend rapidly towards plateaus which are close to the
537 reference values for large S . These asymptotic values depend on the micromechanical model,
538 i.e., the anisotropic or isotropic model used to calculate the mean values \bar{K} and \bar{G} . Second, the
539 dispersion of the results decreases again with an increasing subdomain size. In addition, Figures
540 9a and 9b show that whatever the micromechanical used, the mean values \bar{K} and their associated
541 dispersion (through standard deviation values) are always greater than the mean values \bar{G} and
542 its associated dispersion.

543 The decrease in dispersion with L for both maps evidenced in Figures 9a and 9b can
544 also be illustrated in Figures 10a and 10b, which display the evolutions of the variance of elastic
545 moduli as a function of the subdomains area S (i.e., L^2). Figures 10a and 10b also show that the
546 power fit introduced by equation (3) is a relevant model to quantitatively describe these

547 dispersion evolutions and, hence, to determine the integral range; the calculated regression
548 coefficient R^2 values are all greater than 0.99. For each map, the parameters of these fits,
549 especially the exponents are very close (COx map: 0.89 to 0.896 for the bulk modulus and 0.983
550 to 1.026 to the shear modulus; Toar map: 0.83 for the bulk modulus and 0.813 to 0.824 to the
551 shear modulus), which illustrates the fact that the calculated variances are of the same order of
552 magnitude for any micromechanical model and any elastic modulus under study. Moreover,
553 Figure 10b shows that the variances of the elastic moduli of the Toar map are almost
554 independent of the chosen micromechanical model.

555 Figures 11a and 11b show the mechanical L_{REA} values inferred from the calculations of
556 integral range values obtained from Figures 10a and 10b and equation (8). These figures
557 highlight at least three results. First, in the case of both mineral maps under study, these figures
558 show that for any micromechanical model and any elastic modulus under consideration, the
559 mechanical L_{REA} is significantly greater than morphological L_{REA} (compare Figures 11a and 11b
560 to Figure 8). The concept of L_{REA} fundamentally depends on the property of interest. Second,
561 all things being equal, the mechanical L_{REA} estimates of the COx map are lower than those of
562 the Toar map (see also Table 6). This hierarchy previously observed for the morphological L_{REA}
563 estimates (see Figure 8) is retrieved when the mechanical properties are considered. Third, for
564 any mineral map and any micromechanical model, the L_{REA} estimates of the bulk modulus are
565 lower than those of the shear modulus (Table 6). However, this difference between both
566 estimates is more tenuous in the case of the Toar map for which L_{REA} estimates for a given ε
567 value can be considered as similar and insensitive to the chosen micromechanical models in a
568 first order approach (Table 6).

569 *4.3 Impacts of anisotropy and heterogeneity at the map scale*

570 The calculations of different L_{REA} in this study are based on equations (7) and (8), which
571 *a priori* assume an isotropy of mechanical properties of interest. In this framework, the well-

572 known anisotropic nature of shale raises a difficulty that has been bypassed here by
573 “isotropizing” the transverse isotropic stiffness tensor of the clay matrix. Moreover, to study
574 the impact of this underlying anisotropy introduced in this manner, a pure isotropic
575 micromechanical model has been introduced for comparison purposes. Thus, the main objective
576 associated with the introduction of an implicit anisotropic model and a pure isotropic model
577 was to address the following question: what is the impact of the underlying mechanical
578 anisotropy of shale on its L_{REA} estimates?

579 All the outcomes provided by this work show that the following qualitative results do
580 not depend finally on the chosen micromechanical models and, thus, would be independent of
581 the underlying anisotropic nature of shale. These outcomes are as follows: (i) The mechanical
582 L_{REA} estimates are greater than the morphological ones; (ii) The mechanical L_{REA} estimates of
583 the Toar map are greater than those of the COx map; and (iii) The mechanical L_{REA} estimates
584 of the apparent shear modulus are greater than those of the bulk modulus.

585 The second underlying assumption used in this work is the statistical homogeneity of
586 both mineral maps, meaning, from a statistical viewpoint, that the stationarity assumption of
587 the properties of interest is satisfied on both maps (see appendix and Lantuéjoul, 1991;
588 Torquato, 2013). Let us recall that the stationary assumption means here that the statistical
589 properties implied in our calculations are invariant by translation, i.e., they do not depend on
590 the absolute position in the mineral maps. In our case, the L_{REA} estimated by the CB method
591 strongly depends on the starting domain chosen in the mineral maps, as shown by the high
592 values of standard deviation of the calculated mean L_{REA} values (Table 4). This strong spatial
593 variability of the L_{REA} estimate is a serious indication that the stationarity assumption would not
594 be fulfilled. This indication seems to be confirmed at least for the Toar map by the statistical
595 approach promoted in this study. Figure 12 clearly shows that the 1/S fit associated with
596 equation (2) is finally in poor agreement with the calculated pseudovariance of the clay fraction

597 used to calculate the integral range A_2 (coefficient of determination R^2 equal to 0.36).
598 According to Lantuéjoul (1991), a poor agreement between equation (2) and the measurements
599 would indicate that the image under study would not satisfy the stationary assumption. This
600 result suggests that a combined use of the simple CB method and a calculation of the integral
601 range following the statistical approach applied here would be a simple and efficient way to
602 check *a posteriori* the statistical homogeneity of the maps and images under study at the
603 mesoscopic scale, which is scarcely ensured in practice. This suggestion and these results have
604 to be obviously verified on other mineral maps.

605 **5. Concluding remarks**

606 The main objective of this work was to provide new estimates of REA sizes of the
607 following two shales actively studied in the framework of the deep disposal of radioactive
608 waste: Callovo-Oxfordian (COx) claystone from the Meuse/Haute-Marne underground
609 research laboratory (Eastern France) and Toarcian argillite from the experimental station of
610 Tournemire (Southern France). The L_{REA} estimates obtained from two mineral maps at a
611 mesoscopic scale have been calculated by the classic counting box (CB) method and a
612 statistical approach that introduces the concept of a “statistical” REA. Following this approach,
613 a “statistical” REA is related not only to the microstructure and the properties of each of the
614 components but, above all, to a given precision in the estimation of the effective property
615 depending on the number of realizations “that one is ready to generate” (Jeulin et al., 2004).
616 The probabilistic concept of realization here is any representation or observation of the
617 microstructure considered with a given size and a given fraction of heterogeneities, i.e., in our
618 case, from a practical viewpoint, a subdomain of a given area of a mineral map. This statistical
619 approach requires the calculations of the apparent elastic moduli in this subdomain or in the
620 set of subdomains, which have been achieved here by using two micromechanical models. The
621 first micromechanical model consisted of an anisotropic inclusion-based model for which

622 spherical nonclay grain is embedded in a clay matrix, in which the values of its transverse
623 isotropic stiffness tensor have been taken from literature. The second micromechanical model
624 was an isotropic inclusion-based model for which spherical nonclay grain is embedded in a
625 clay matrix; the elastic moduli values have been inverted by a Monte-Carlo approach from the
626 macroscopic engineering moduli measured on both shales under study. The calculations
627 performed following this statistical approach have shown the following results:

- 628 • The morphological L_{REA} estimates inferred for the statistical approach are of the
629 same order of magnitude as those found in the literature and obtained by the
630 classic CB method by considering the values of the relative errors, i.e., between
631 5 and 10%.
- 632 • For any micromechanical model and any elastic modulus under consideration,
633 the mechanical L_{REA} is significantly greater than morphological L_{REA} . Our study
634 confirms that the concept of L_{REA} fundamentally depends on the property of
635 interest.
- 636 • The mechanical L_{REA} estimates of the Toar map are greater than those of the COx
637 map. This is also the case for the morphological L_{REA} estimates inferred for a
638 low relative error of 5%.
- 639 • All things being equal, the mechanical L_{REA} estimates of shear modulus are
640 greater than those of the bulk modulus

641 Moreover, our study highlights two additional aspects. First, all the outcomes provided
642 by this work show that these previous qualitative results do not depend on the chosen
643 micromechanical models and, thus, would be ultimately independent of the underlying
644 anisotropic nature of the shale. Second, a combined use of the simple CB method with a
645 calculation of the integral range following this statistical approach strongly brings into question
646 the statistical homogeneity of the Toar map. Consequently, this result suggests that this coupled

647 approach would be a simple and efficient way to check *a posteriori* the statistical homogeneity
648 of the maps and images under study, which is scarcely ensured in practice. The application and
649 validation of such a coupled approach on other mineral maps is obviously one natural
650 perspective of this work.

651 Finally, this work strongly suggests the need for further investigations in two directions.
652 First, this work has considered the definitions of REA or REV initially established on
653 homogeneous and isotropic media, but one may wonder if these definitions are still valid or can
654 be refined for strongly anisotropic media made of anisotropic components. Second, our
655 approach has highlighted the crucial role of the anisotropic mechanical properties of the clay
656 matrix. The accurate measurements of these mechanical parameters are still a challenging area
657 of experimental research.

658 **Appendix. The integral range**

659 Here, we borrow definitions and notations from Lantuéjoul (1991). Let us consider (i) a domain
660 of area S that is sufficiently large to ensure that the properties of interest may be estimated with
661 good precision (note that the following concepts are defined in 2D but their generalization in
662 3D is straightforward), and (ii) a stationary random function $(Z(x))_{x \in \mathbb{R}^d}$ with mean μ and
663 variance $\sigma^2 = D_Z^2$, which are both unknown. In the context of randomness, the average value of
664 Z , named \bar{Z} , over the area S can be evaluated by the following estimator:

$$665 \quad Z(S) = \frac{1}{S} \int_S Z(x) dx \quad (A1)$$

666 and the variance of this estimator, namely, $D_Z^2(S)$, is given by the following:

$$667 \quad D_Z^2(S) = \frac{1}{S^2} \int_S \int_S C(y-x) dx dy \quad (A2)$$

668 where $C(y-x)$ is the autocovariance function of the *stationary* random function $Z(x)$
669 expressed by the following:

670 $C(y-x) = C(h) = E\{[Z(x) - E(Z)][Z(x+h) - E(Z)]\}$ (A3)

671 In other words, the statistical property $D_Z^2(S)$ defined by equation (A2) can be seen as the
 672 variance of the average value $\bar{Z}(S)$, which is in fact the effective property to be determined. As
 673 explained further, this statistical property $D_Z^2(S)$ is directly linked to the concept of the integral
 674 range. The next step requires the definition of the ergodicity assumption, as follows: a random
 675 function Z is ergodic if variance $D_Z^2(S)$ tends toward 0 when the size of the domain S becomes
 676 infinite, as follows:

677 $\lim_{S \rightarrow \infty} D_Z^2(S) = 0$ (A4)

678 The concept of the integral range, namely, A_2 , is then introduced to define the rate of decrease
 679 of this variance at large distances, i.e., in large areas, as follows:

680 $A_2 = \lim_{S \rightarrow \infty} S \frac{D_Z^2(S)}{D_Z^2}$ (A5)

681 This quantity does not always exist. When it does exist, it is nonnegative. If $A < +\infty$, for large
 682 S , then:

683 $D_Z^2(S) \approx \frac{D_Z^2 A_2}{S}$ (A6)

684 Now we return to the problem met in practice. Suppose that the domain S can be decomposed
 685 into a union of disjoint subdomains, s_1, \dots, s_k , all of the same shape and of the same size s . In
 686 each subdomain s_i , the average value taken by $Z(x)$ over s_i is as follows:

687 $Z(s_i) = \frac{1}{s} \int_{s_i} Z(x) dx$ (A7)

688 where s denotes the area of s_i . The “dispersion variance” of the $Z(s_i)$ is now defined as follows:

689 $\sigma^2(s|S) = E \left\{ \frac{1}{k} \sum_{i=1}^k [Z(s_i) - Z(S)]^2 \right\}$ (A8)

690 or

691 $\sigma^2(s|S) = D_Z^2(s) - D_Z^2(S)$ (A9)

692 Suppose now that the integral range is finite and nonzero and that the area s is very large
693 compared to A_2 . In this case, the approximation formula (A6) is valid and then equation (A9)
694 can be rewritten as follows:

$$695 \quad \sigma^2(s|S) \approx D_Z^2 A_2 \left(\frac{1}{s} - \frac{1}{S} \right) \quad (A10)$$

696 Now if S has been divided into a large number of subdomains, then $1/S$ can be considered as
697 negligible compared to $1/s$ and therefore:

$$698 \quad \sigma^2(s|S) \approx \frac{D_Z^2 A_2}{s} \quad (A11)$$

699 Thus, the dispersion variance is inversely proportional to the area of the subdomains, s . From a
700 practical viewpoint, it is this dispersion variance that is calculated to obtain an estimation of A_2
701 by a fitting procedure.

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708

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887 **Table captions**

888 Table 1. *Geometrical and mineralogical features of both mineral maps used in this work.*

889 Table 2. Input data for the calculations of apparent elastic properties of both shales under study.

890 Table 3. Inverted values of isotropic moduli of the clay phase by the Monte Carle procedure
891 (isotropic model).

892 Table 4. REA estimates obtained from the counting box method.

893 Table 5. Morphological REA size estimates by statistical approach (n=1; error ranging between
894 5 and 10%). The results of inverse regression and integral range are also given.

895 Table 6. Recapitulation of mechanical REA size estimates by statistical approach (n=1; error
896 ranging between 5 and 10%).

897

898 **Figure captions**

899 Figure 1. Microstructure of clay-rocks at various scales (modified from Ulm et al. 2005; Sarout
900 and Guéguen, 2008; Cosenza et al., 2015a).

901 Figure 2. Extraction of the mineral COx map used in this work (modified from Cosenza et al.,
902 2015a).

903 Figure 3. Extraction of the mineral Toar map used in this work (modified from Fauchille, 2015).

904 Figure 4. A. Partitioning of the COx map only used for the counting box method. B. Partitioning
905 of the Toar map used for the counting box method. In both cases, the direction of
906 bedding is indicated.

907 Figure 5 Estimation of the Representative Elementary Area (REA) size of mineral maps by
908 counting box method. Evolution of mean clay fraction with increasing subdomain size
909 (box) and for different starting domains. The coordinates of starting domains are given
910 in the captions boxes (see the origin of the system of Cartesian coordinates in Figures 2
911 and 3).

912 a. COx map. The x-coordinates of the starting domains is 250 μm . The horizontal dashed
913 lines indicate the range of mean clay fraction corresponding to $(1 \pm \varepsilon)\bar{\phi}_{COx}$ with
914 $\bar{\phi}_{COx}=0.504$ (50.4%) and $\varepsilon=0.1$ (10%)

915 b. Toar map. The horizontal dashed lines indicate the range [62.9-76.9%] corresponding
916 to $(1 \pm \varepsilon)\bar{\phi}_{Toar}$ with $\bar{\phi}_{Toar}=0.699$ (69.9%) and $\varepsilon=0.1$ (10%).

917 Figure 6. Evolution of the mean clay fraction and related standard deviation *versus* the
918 subdomain size for both maps.

919 Figure 7. Determination of the “morphological” integral range A_2 . Evolution of the
920 pseudovariance of the clay fraction, i.e., Variance ($\bar{\varphi}_{arg}(1-\bar{\varphi}_{arg})$) as a function of $1/S$,
921 where S is the box size area. A linear fit is indicated by a bold line. a. COx map. b. Toar
922 map.

923 Figure 8. REA size, L_{REA} (μm), as a function of error (%) for one realization ($n=1$) for both
924 mineral maps.

925 Figure 9. Mean value and dispersion (standard deviation) of the apparent elastic moduli as a
926 function of the subdomain size L in the case of the anisotropic model and isotropic
927 model. a: COx map. b: Toar map.

928 Figure 10. Determination of the “mechanical” integral range A_2 . Evolution of the variance of
929 the elastic modulus of the anisotropic and isotropic models as a function of the
930 subdomain area. Power fits are also indicated to calculate the integral range values. a.
931 COx map. b. Toar map.

932 Figure 11. Mechanical REA size, L_{REA} (μm), as a function of error (%) for one realization ($n=1$).
933 Both elastic moduli, i.e., the bulk modulus and shear modulus; both micromechanical
934 models, i.e., the anisotropic model and isotropic model, are considered. a. COx map. b.
935 Toar map.

936 Figure 12. Zoom of the evolutions of the pseudo-variance of clay fraction i.e., Variance($\bar{\varphi}_{arg}(1-$
937 $\bar{\varphi}_{arg})$) as a function of subdomain area of both maps. R^2 values of both fits are also
938 given.

939

940

941 Table 1. *Geometrical and mineralogical features of both mineral maps used in this work.*

Map	Resolution (μm)	Total number of pixels	Dimensions (pixels)	Dimensions (μm)	Clay minerals (%)	Quartz (%)	Calcite (%)
COx	2	384 000	250 x 1536	500 x 3072	50.4	13.8	25.0
Toar	0.625	$16 \cdot 10^6$	4000 x 4000	2500 x 2500	69.9	13.2	14.1

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944

945 Table 2. Input data for the calculations of apparent elastic properties of both shales under study.

Shale	Macroscopic (reference) shale elastic modulus (GPa)	Non-clay phase Isotropic elastic modulus Weighted average (GPa) ^b	Clay phase Transverse isotropic (undrained) modulus (GPa) ^c
Callovo-Oxfordian claystone	Bulk modulus ^a	Bulk modulus	C ₁₁ = 14.3
	11.5	56.3	C ₁₂ =4.6
	Shear modulus ^a	Shear modulus	C ₁₃ =2.7
	5.3	38.5	C ₃₃ =8.9 C ₄₄ =2.8
Tournemire argillite	Bulk modulus ^d	Bulk modulus	C ₁₁ = 14.3
	9.2	53.9	C ₁₂ =4.6
	Shear modulus ^d	Shear modulus	C ₁₃ =2.7
	6.5	37.5	C ₃₃ =8.9 C ₄₄ =2.8

946 ^aANDRA (2009) (undrained Poisson ratio: $\nu^u=0.3$)

947 ^bTable 1 and Mavko et al. (2009)

948 ^cUlm et al. (2005); Cosenza et al. (2015b)

949 ^dNiandou et al. (1997)

950

951 Table 3. Inverted values of isotropic moduli of clay phase by Monte Carlo procedure (isotropic
952 model).

Shale	Clay phase Inverted isotropic modulus (GPa)	Relative difference between reference value and calculated value from inverted clay phase moduli (%)
Callovo-Oxfordian claystone	Bulk modulus	Bulk modulus
	5.0	0.3
Tournemire argillite	Shear modulus	Shear modulus
	1.6	1.4
Tournemire argillite	Bulk modulus	Bulk modulus
	5.3	1.6
Tournemire argillite	Shear modulus	Shear modulus
	3.7	0.1

953

954

955 Table 4. REA estimates obtained by counting box method.

956

Map	Area (A_i)	Center of area (C_i)	Asymptotic value (φ_∞) (%)	REA estimate (L_{REA}) (μm) $\varepsilon=0.1$ (10%)	REA estimate (L_{REA}) (μm) $\varepsilon=0.05$ (5%)
COx	A1	C1 (x=250 μm , z=250 μm)	49.0	262	314
	A2	C2 (x=250 μm , z=750 μm)	48.6	322	438
	A3	C3 (x=250 μm , z=1250 μm)	49.4	128	170
	A4	C4 (x=250 μm , z=1750 μm)	51.6	72	86
	A5	C5 (x=250 μm , z=2250 μm)	52.4	202	310
	A6	C6 (x=250 μm , z=2500 μm)	51.5	50	86
			Mean: 51.5	Mean: 172.7	Mean: 234.0
			St. Dev.: 1.6	St. Dev.: 108.0	St. Dev.: 142.6
Toar	A1	C1 (x=625 μm , z=625 μm)	71.4	163	179
	A2	C2 (x=1250 μm , z=625 μm)	68.5	71	749
	A3	C3 (x=625 μm , z=1250 μm)	70.2	214	260
	A4	C4 (x=1250 μm , z=1250 μm)	69.1	68	576
			Mean: 69.8	Mean: 129.0	Mean: 441.0
			St. Dev.: 1.3	St. Dev.: 71.8	St. Dev.: 267.4

957

958 Table 5. Morphological REA size estimates by statistical approach (n=1; error ranging between 5 and 10%). The results of inverse regression and
 959 integral range are also given.

Shale	Statistical morphological REA		
	Regression $\frac{D_{\varphi}^2(S)}{\bar{\varphi}(1-\bar{\varphi})} = \frac{A_2}{S}$	Integral range A ₂	REA size estimates (μm) ($\varepsilon \in [5,10]\%$)
Callovo-Oxfordian claystone	$\frac{D_{\varphi}^2(S)}{\bar{\varphi}(1-\bar{\varphi})} = \frac{106.6}{S}$ R ² = 0.96	106.6	205-410
Tournemire argillite	$\frac{D_{\varphi}^2(S)}{\bar{\varphi}(1-\bar{\varphi})} = \frac{703}{S}$ R ² = 0.36	703	345-696

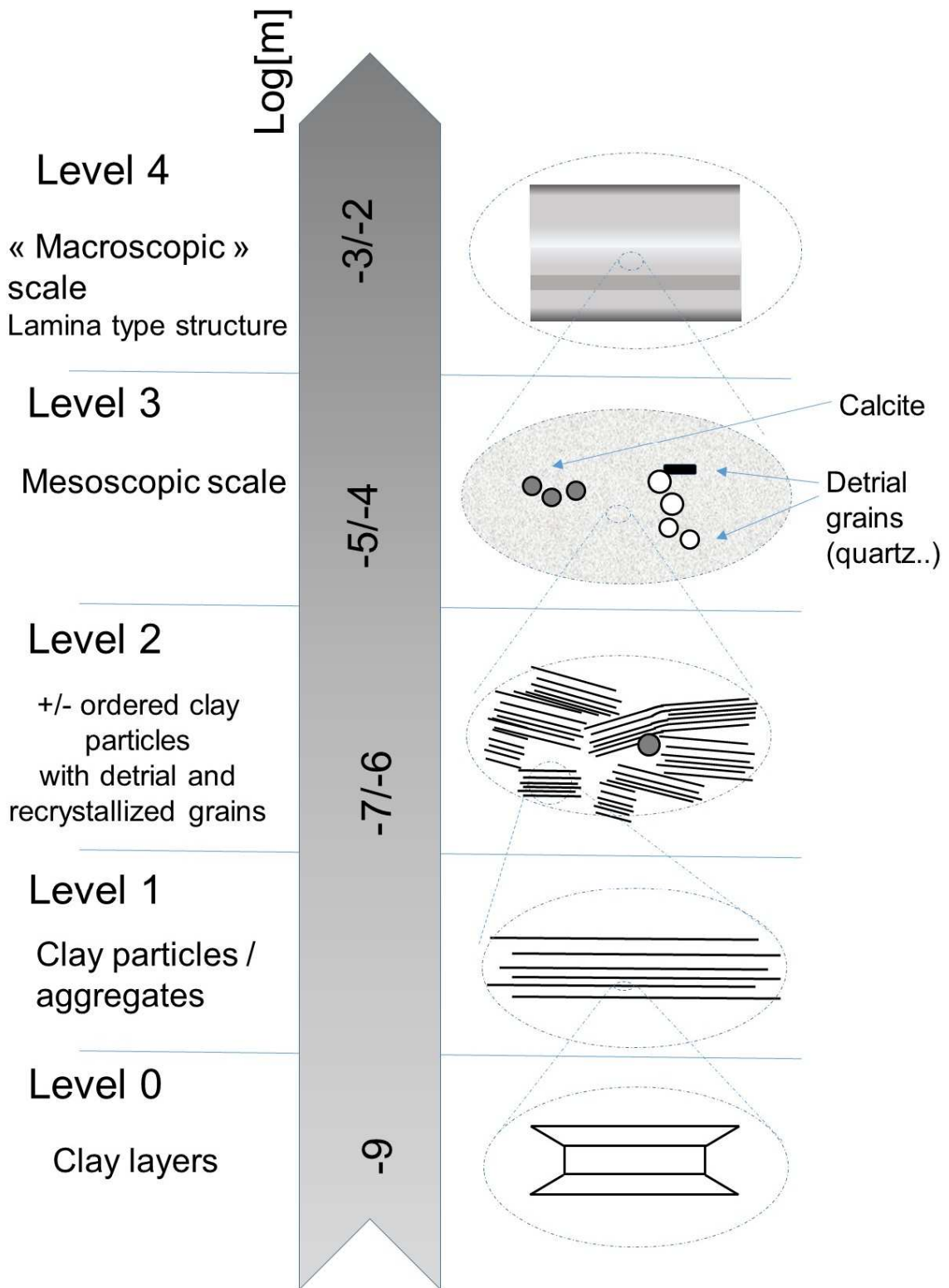
960

961 Table 6. Recapitulation of mechanical REA size estimates by statistical approach (n=1; error ranging between 5 and 10%).

Shale	Statistical mechanical REA							
	Anisotropic model				Isotropic model			
	Bulk modulus		Shear modulus		Bulk modulus		Shear modulus	
Integral range A ₂	REA size estimates (μm) $\varepsilon \in [5,10]\%$	Integral range A ₂	REA size estimates (μm) $\varepsilon \in [5,10]\%$	Integral range A ₂	REA size estimates (μm) $\varepsilon \in [5,10]\%$	Integral range A ₂	REA size estimates (μm) $\varepsilon \in [5,10]\%$	
Callovo-Oxfordian claystone	27.5	353-765	27.7	574-1251	28.2	253-512	29.4	338-664
Tournemire argillite	37.4	743-1759	37.9	748-1772	38.4	687-1594	38.3	720-1688

962

963 Figure 1. Microstructure of clay-rocks at various scales (modified from Ulm et al. 2005; Sarout
 964 and Guéguen, 2008; Cosenza et al., 2015a).

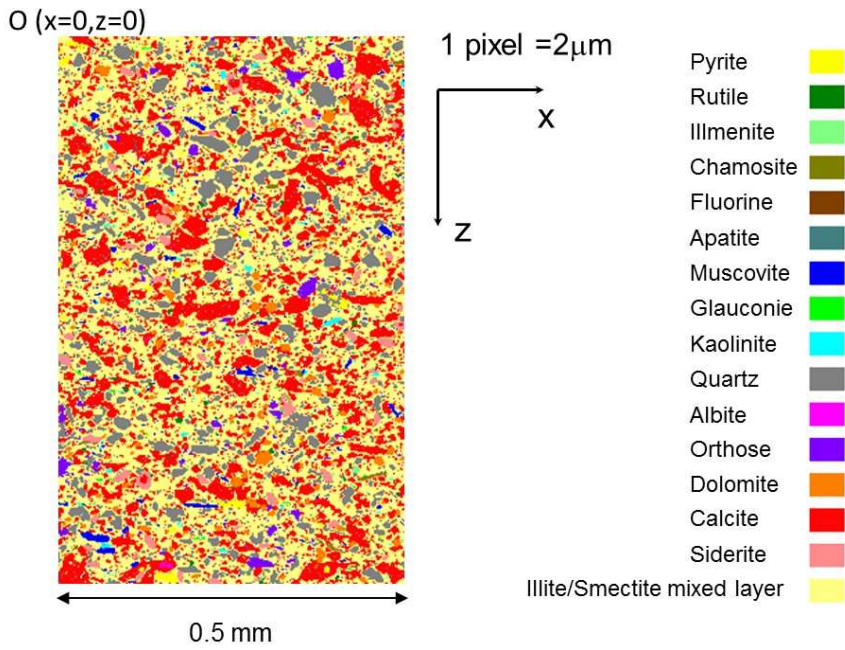


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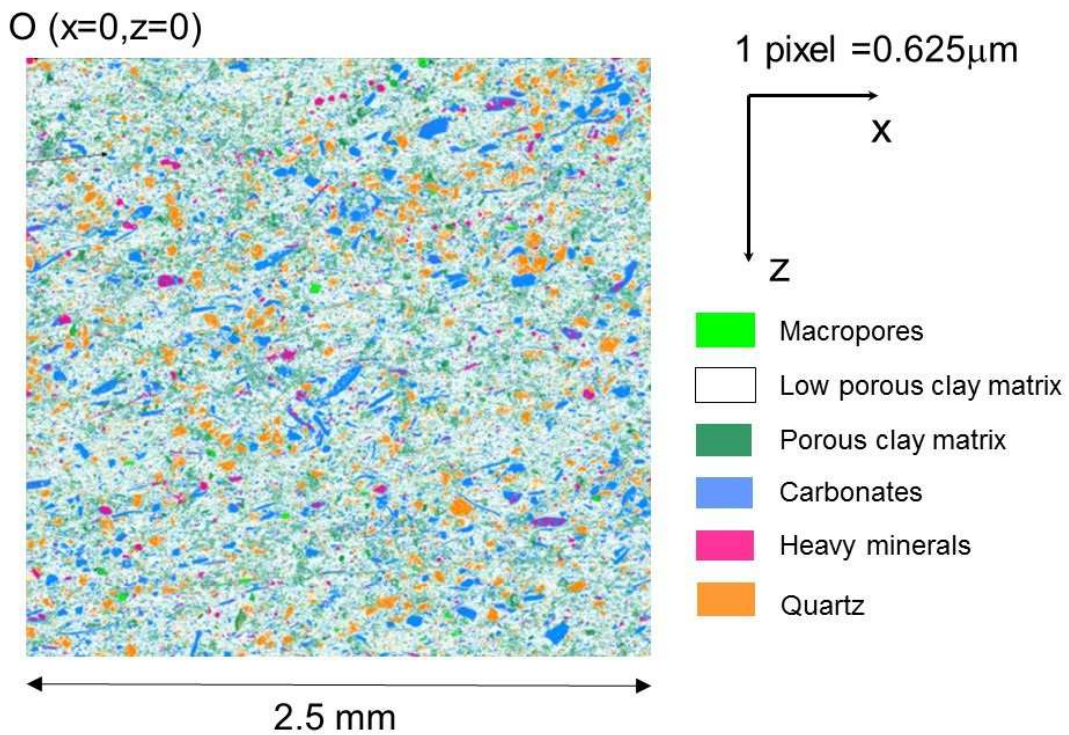
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970 Figure 2. Extraction of the mineral COx map used in this work (modified from Jorand, 2006).

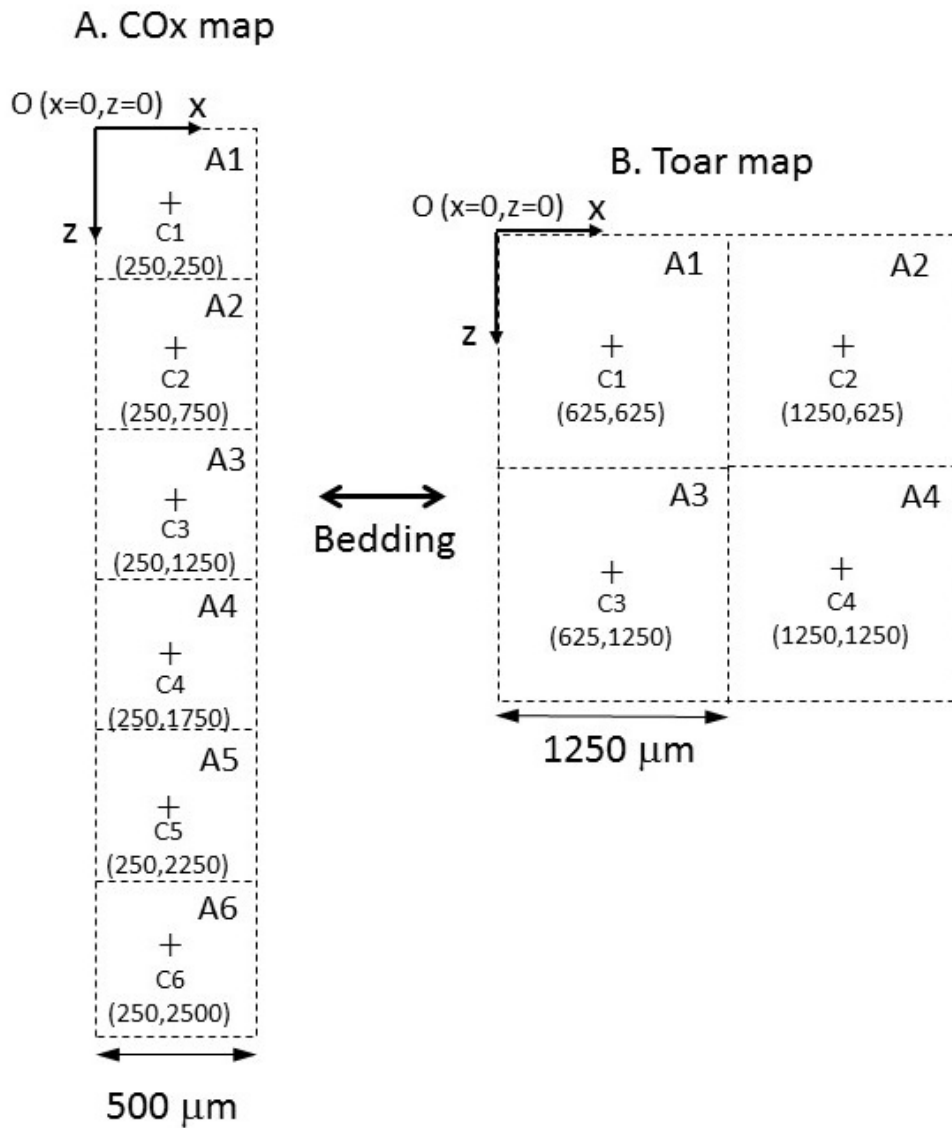
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973 Figure 3. The mineral Toar map used in this work (modified from Fauchille, 2015).

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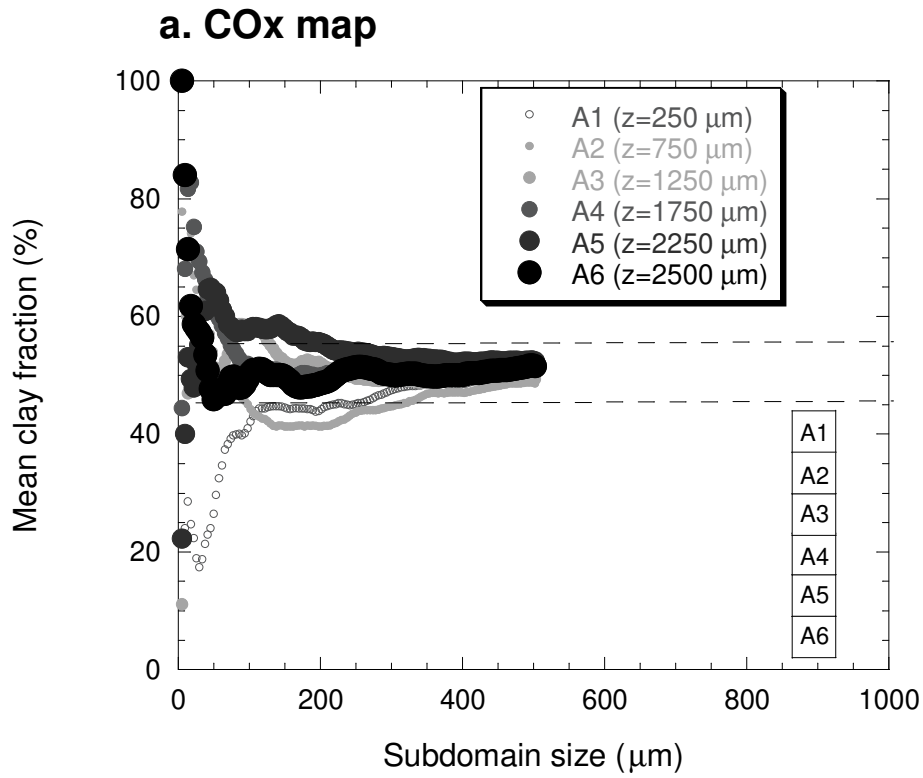
977 Figure 4. A. Partitioning of the COx map only used for counting box method. B. Partitioning

978 of the Toar map used for counting box method. In both cases, the direction of bedding

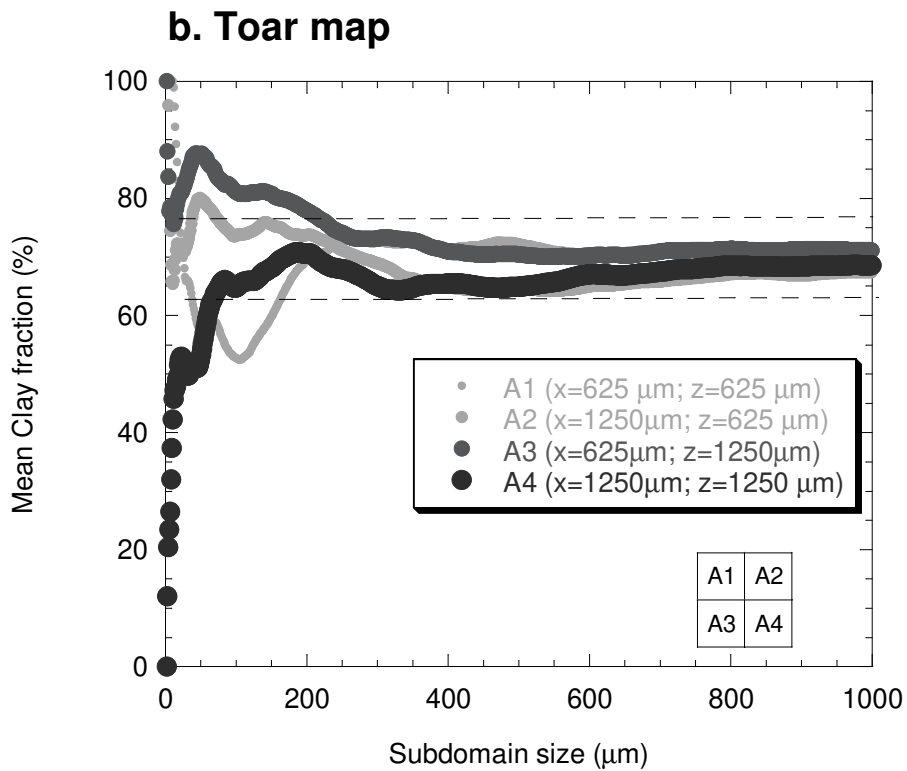
979 is indicated.

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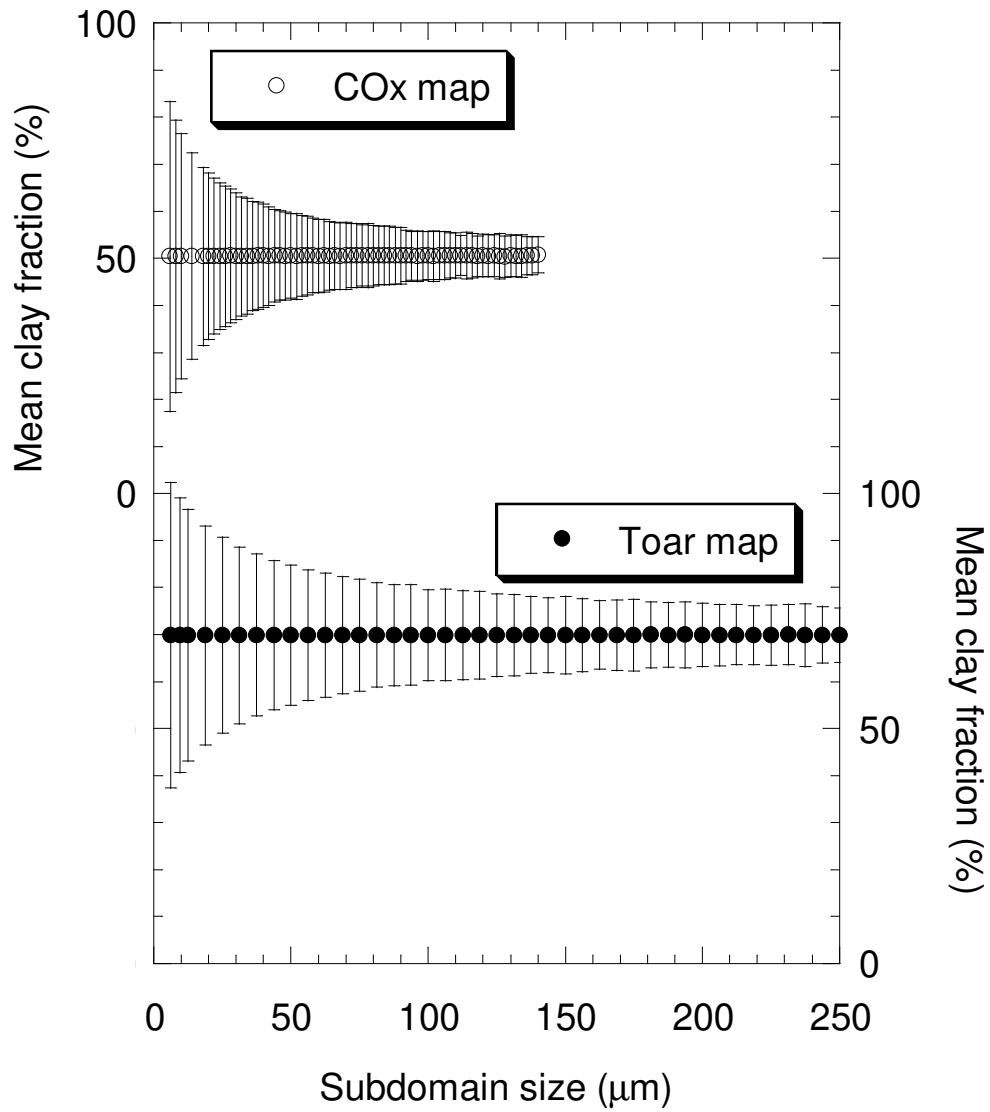
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983

984 Figure 5. Estimation of the Representative Elementary Area (REA) size of mineral maps by
 985 counting box method. Evolution of mean clay fraction with increasing subdomain size
 986 (box) and for different starting domains. The coordinates of starting domains are given
 987 in the captions boxes (see the origin of the system of Cartesian coordinates in Figures 2
 988 and 3).

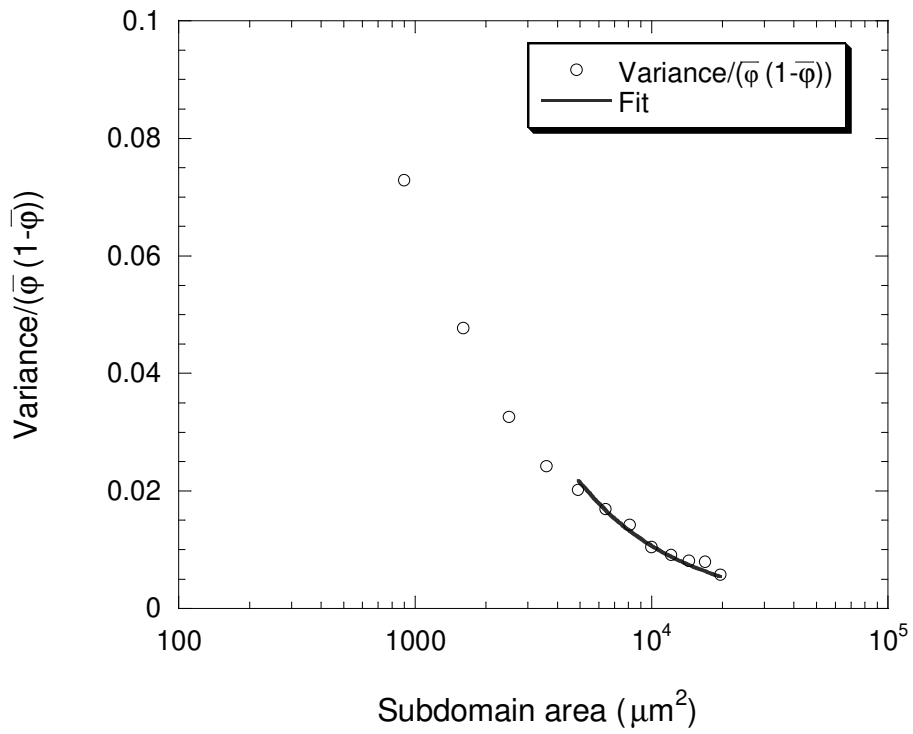
- 989 a. COx map. The x-coordinates of the starting domains is 250 μm . The horizontal dashed lines
990 indicate the range of mean clay fraction corresponding to $(1 \pm \varepsilon)\bar{\phi}_{COx}$ with $\bar{\phi}_{COx}=0.504$
991 (50.4%) and $\varepsilon=0.1(10\%)$.
- 992 b. Toar map. The horizontal dashed lines indicate the range [62.9-76.9%] corresponding to
993 $(1 \pm \varepsilon)\bar{\phi}_{Toar}$ with $\bar{\phi}_{Toar}=0.699$ (69.9%) and $\varepsilon=0.1(10\%)$.
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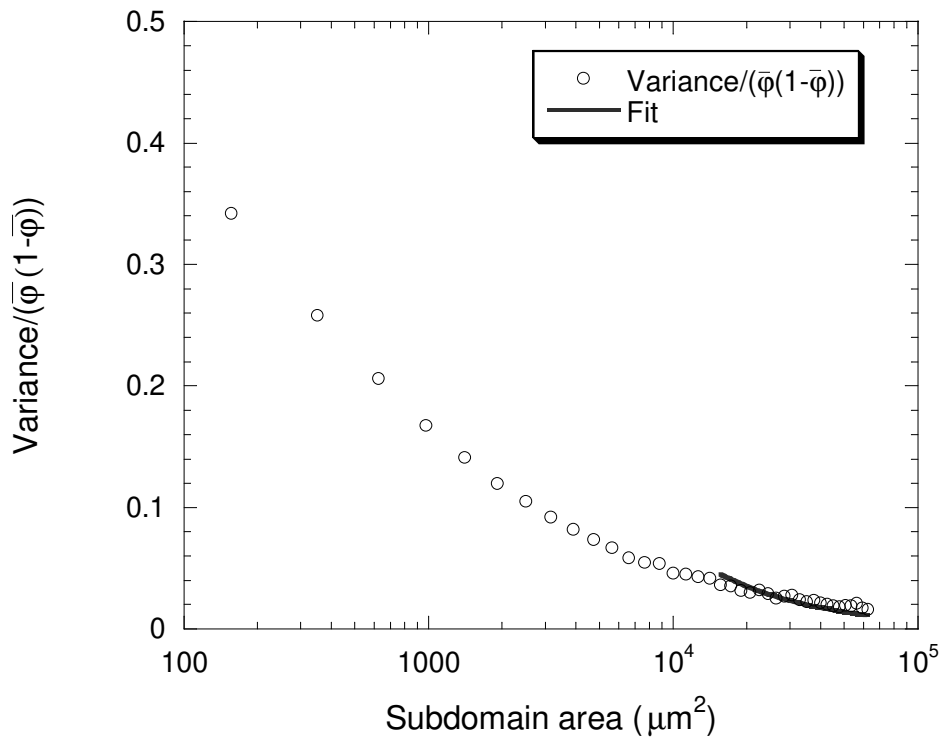
Figure 6. Evolution of the mean clay fraction and related standard deviation versus the subdomain size L for both maps.

a. COx map



1000

b. Toar map

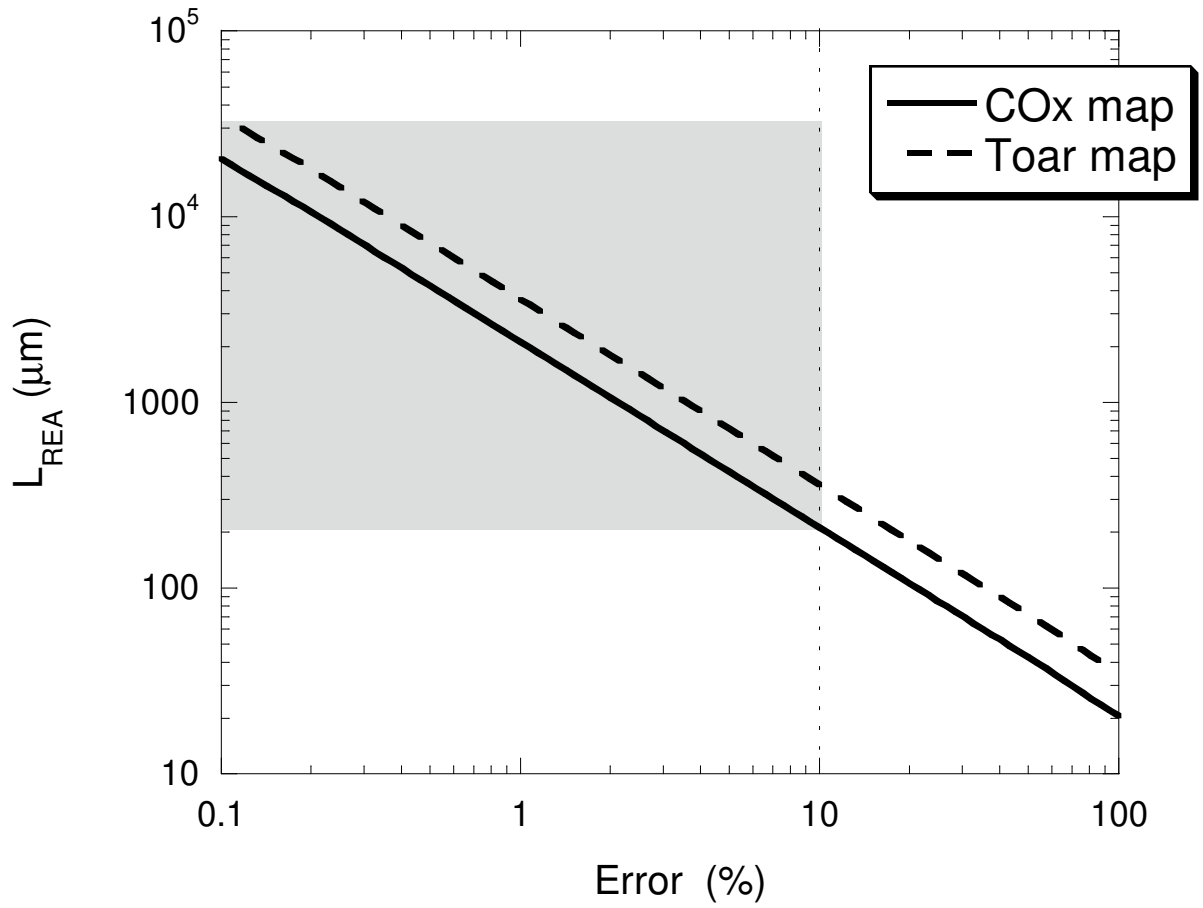


1001

1002 Figure 7. Determination of the “morphological” integral range A_2 . Evolution of the pseudovariance of
1003 the clay fraction, i.e., $\text{Variance}(\bar{\varphi}_{arg}(1-\bar{\varphi}_{arg}))$ as a function of $1/S$, where S is the box size area.

1004 A linear fit is indicated by a bold line. a. COx map. b. Toar map.

1005

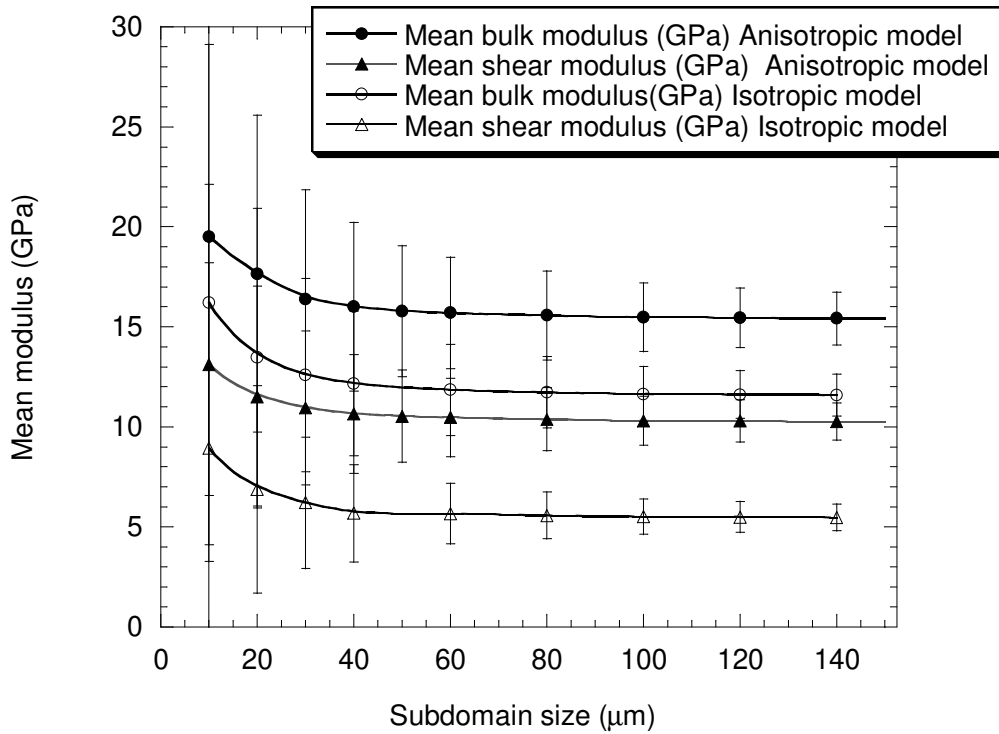


1006

1007 Figure 8. Morphological REA size, L_{REA} (μm) as a function of error (%) for one realization
 1008 (n=1) for both mineral maps.

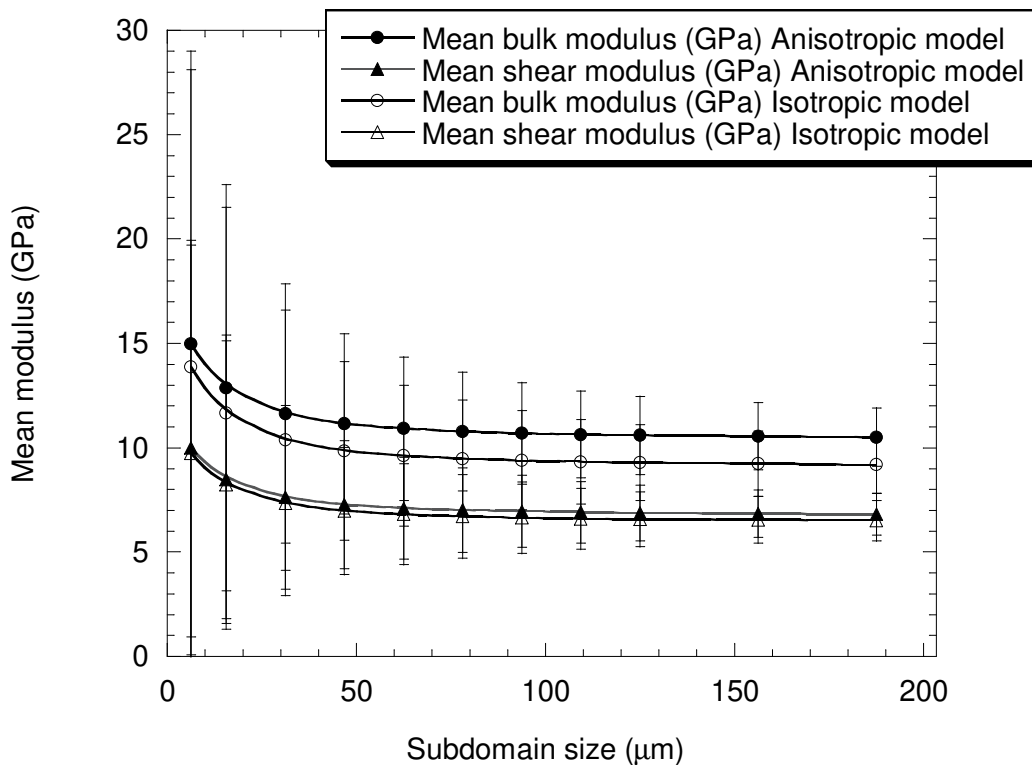
1009

a. COx map



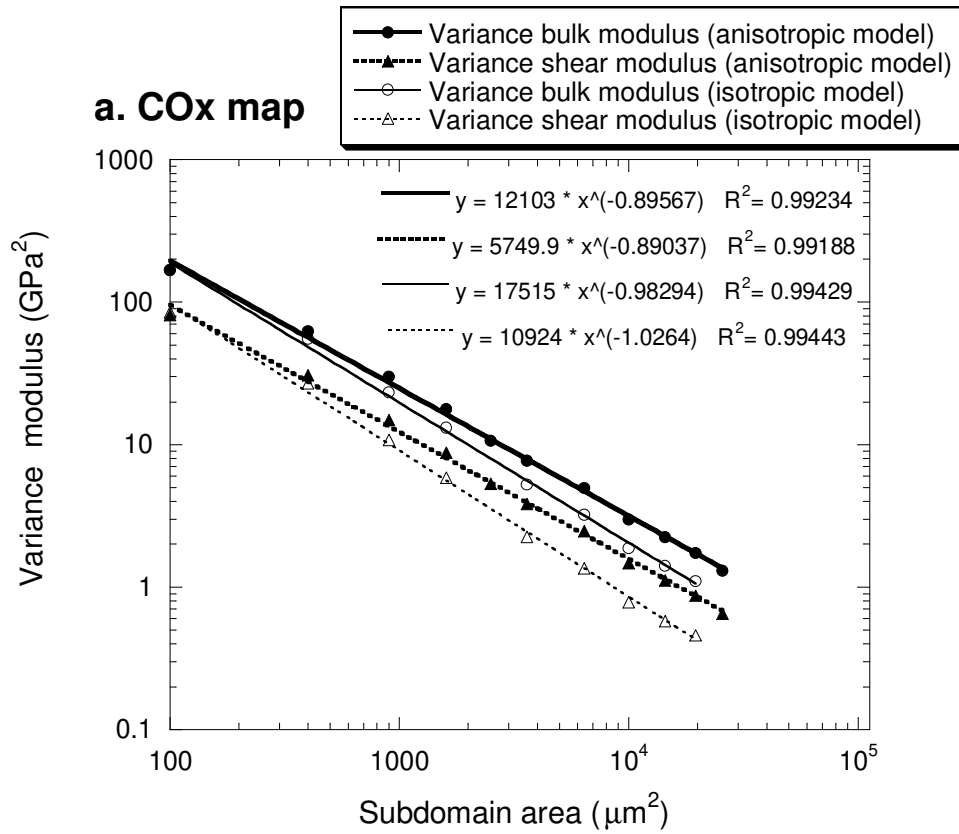
1010

b. Toar map

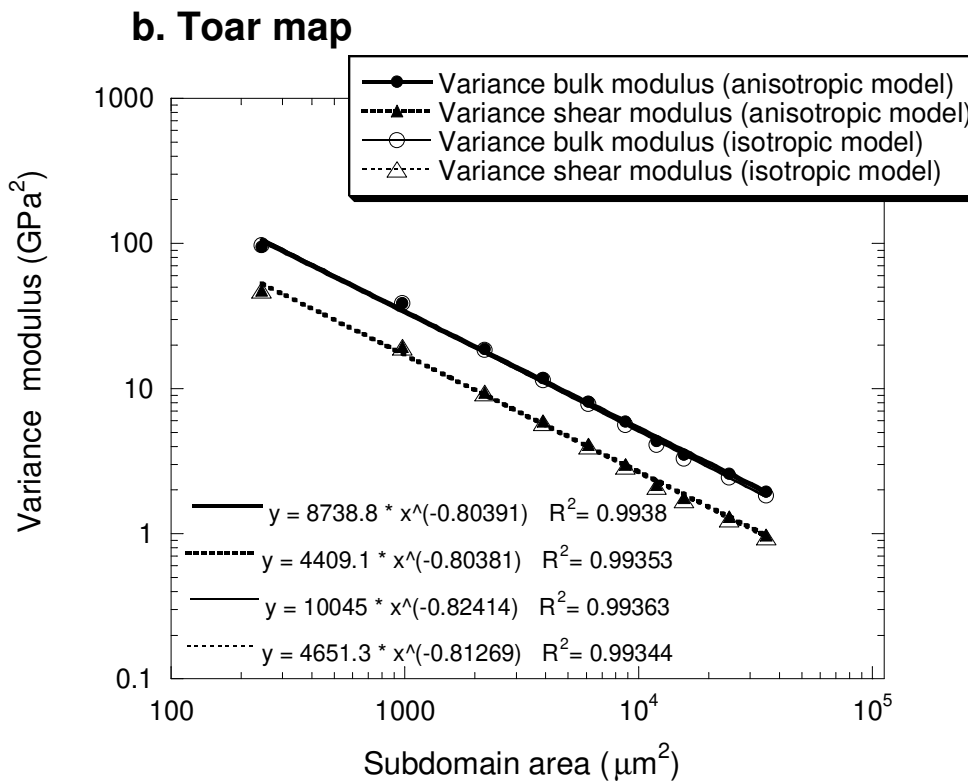


1011

1012 Figure 9. Mean value and dispersion (standard deviation) of the apparent elastic moduli as a
 1013 function of the subdomain size L in the case of the anisotropic model and isotropic
 1014 model. a: COx map. b: Toar map.



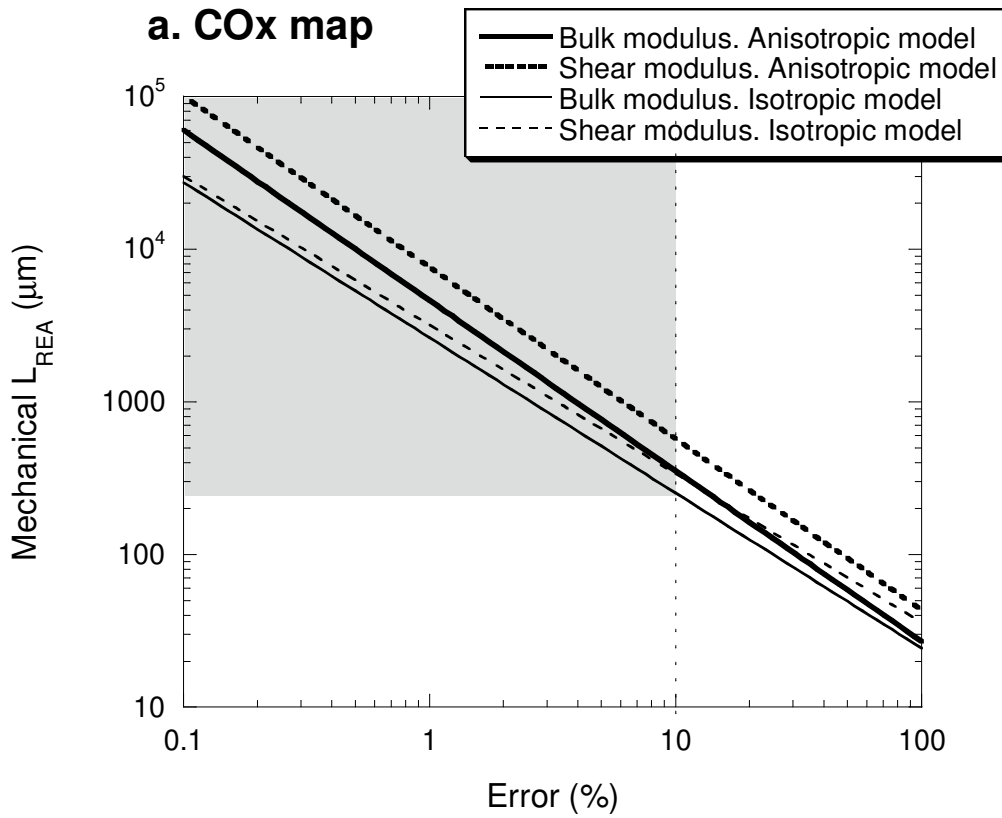
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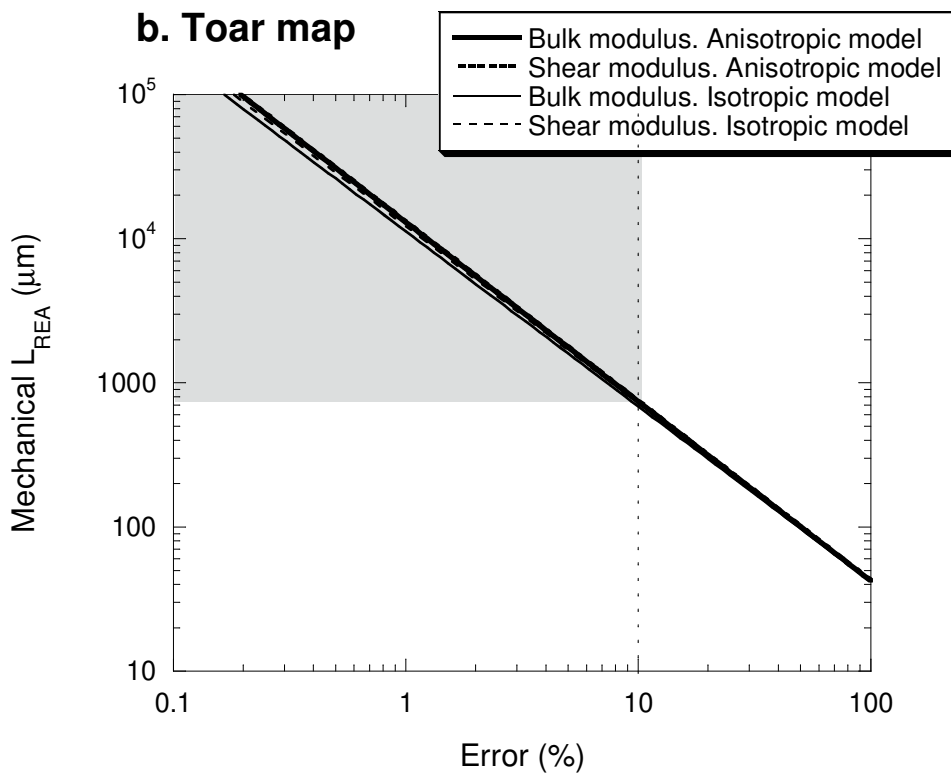
1017 Figure 10. Determination of the “mechanical” integral range A_2 . Evolution of the variance of the elastic
 1018 modulus of the anisotropic and isotropic models as a function of the subdomain area. Power fits
 1019 are also indicated to calculate the integral range values. a. COx map. b. Toar map.

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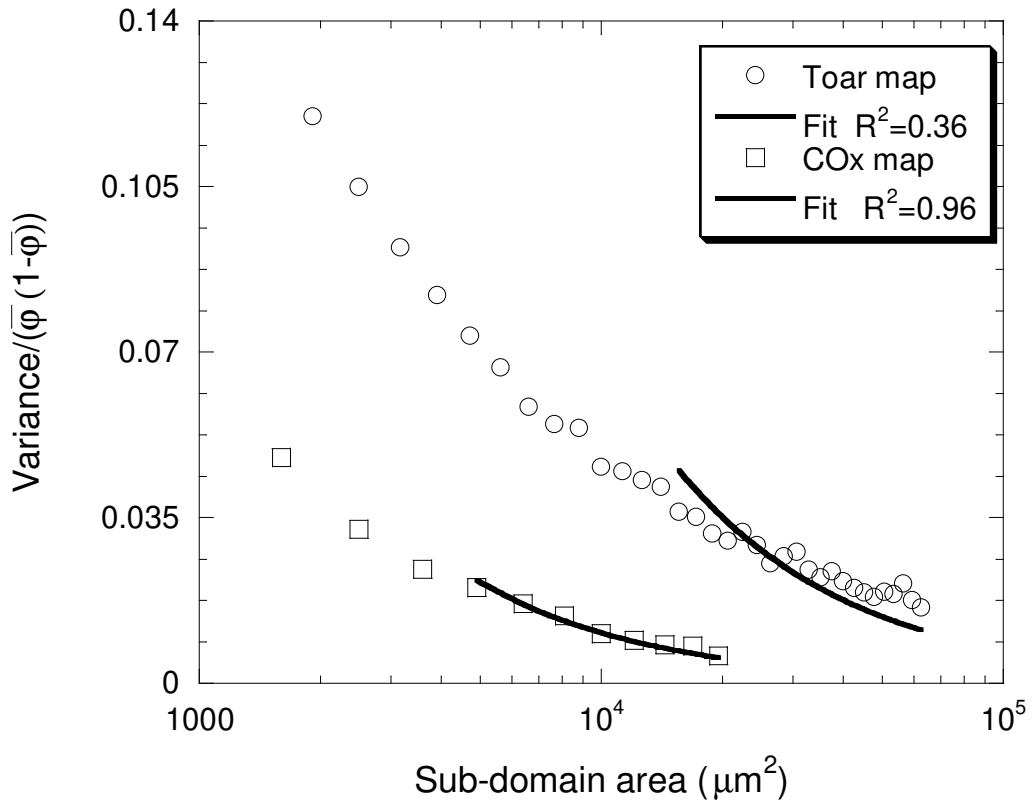
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1023

1024 Figure 11. Mechanical REA size, L_{REA} (μm), as a function of error (%) for one realization ($n=1$). Both
 1025 elastic moduli, i.e., the bulk modulus and shear modulus; both micromechanical models, i.e.,
 1026 the anisotropic model and isotropic model, are considered. a. COx map. b. Toar map.



1027

1028 Figure 12. Zoom of the evolutions of the pseudo-variance of clay fraction i.e., $\text{Variance}(\bar{\varphi}_{arg}(1-$
 1029 $\bar{\varphi}_{arg}))$ as a function of subdomain area of both maps. R^2 values of both fits are also
 1030 given.

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