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Quantification of the pore size distribution of soils: Assessment of existing software using tomographic and synthetic 3D images.

A. N. Houston^a, W. Otten^{a,b}, R. Falconer^{a,c}, O. Monga^d, P.C.Baveye^e, S.M. Hapca^{a,f*}

^a School of Science, Engineering and Technology, Abertay University, 40 Bell Street, Dundee DD1 1HG, UK.

^b current address: School of Water, Energy and Environment, Cranfield University, Cranfield MK43 0AL, UK.

^c current address: School of Arts, Media and Computer Games , Abertay University, 40 Bell Street, Dundee DD1 1HG, UK.

^d UMMISCO-Cameroun, UMI 209 UMMISCO, University of Yaoundé, IRD, University of Paris 6, F-93143 Bondy Cedex, France.

^e UMR ECOSYS, AgroParisTech, Université Paris-Saclay, Avenue Lucien Brétignières, Thiverval-Grignon 78850, France.

^f current address: Dundee Epidemiology and Biostatistics Unit, School of Medicine, Dundee University, Kirsty Semple Way, Dundee DD2 4BF, UK.

* Corresponding author. E-mail: s.z.hapca@dundee.ac.uk

23 **Abstract:**

24

25 The pore size distribution (PSD) of the void space is widely used to predict a range of
26 processes in soils. Recent advances in X-ray computed tomography (CT) now afford novel
27 ways to obtain exact data on pore geometry, which has stimulated the development of
28 algorithms to estimate the pore size distribution from 3D data sets. To date there is however
29 no clear consensus on how PSDs should be estimated, and in what form PSDs are best
30 presented. In this article, we first review the theoretical principles shared by the various
31 methods for PSD estimation. Then we select methods that are widely adopted in soil science
32 and geoscience, and we use a robust statistical method to compare their application to
33 synthetic image samples, for which analytical solutions of PSDs are available, and X-ray CT
34 images of soil samples selected from different treatments to obtain wide ranging PSDs.
35 Results indicate that, when applied to the synthetic images, all methods presenting PSDs as
36 pore volume per class size (i.e., Avizo, CTAnalyser, BoneJ, Quantim4, and DTM), perform
37 well. Among them, the methods based on Maximum Inscribed Balls (Bone J, CTAnalyser,
38 Quantim4) also produce similar PSDs for the soil samples, whereas the Delaunay
39 Triangulation Method (DTM) produces larger estimates of the pore volume occupied by
40 small pores, and Avizo yields larger estimates of the pore volume occupied by large pores.
41 By contrast, the methods that calculate PSDs as object population fraction per volume class
42 (Avizo, 3DMA, DFS-FIJI) perform inconsistently on the synthetic images and do not appear
43 well suited to handle the more complex geometries of soils. It is anticipated that the
44 extensive evaluation of method performance carried out in this study, together with the
45 recommendations reached, will be useful to the porous media community to make more
46 informed choices relative to suitable PSD estimation methods, and will help improve current
47 practice, which is often ad hoc and heuristic.

48

49 Keywords: porous media, soil, pore size distribution, computed tomography, X-ray.

50

51 **1. Introduction**

52 In the 1930s and 40s, soil physicists like Haines (1930) and Childs (1940) came to
53 acknowledge that the size distribution of soil particles, routinely measured since the 18th
54 century (Baveye, 2013), provided very little useful information concerning the retention of
55 water and its transport in soils. This realization led to a shift of emphasis from soil particles to
56 the "water-occupied void space [...], which largely determines the gross physical properties
57 of soils" (Childs and Collis-George, 1948). These authors suggested that soil voids, or
58 "pores", could be linked to straight capillaries of varying diameters, and that their size
59 distribution would provide the type of direct quantitative information needed to describe the
60 functioning of soils.

61 This perspective has since become one of the hallmarks of soil physics, and it is adopted
62 in most soil physics textbooks to explain the principles that govern the retention of water in
63 soils and its movement. Thus a significant body of research has been devoted to the use of
64 the pore size distribution (PSD) to predict a wide range of processes of interest, such as gas
65 diffusion, water retention and flow, mechanical resistance, carbon dynamics, microbial
66 colonization, and root penetration (Monga *et al.*, 2009; Pajor *et al.*, 2010; Kravchenko *et al.*,
67 2011a; Falconer *et al.*, 2012; Schmidt *et al.*, 2012; Cazelles *et al.*, 2013; Juarez *et al.*,
68 2013; Zaffar and Lu 2015), as well as to assess the effect of different management practices
69 and degradation processes on soil productivity (Kravchenko *et al.* 2011b, Dal Ferro *et al.*,
70 2012; Muñoz-Ortega *et al.*, 2014; Naveed *et al.*, 2014a; Rab *et al.*, 2014).

71 In parallel with the application of the PSD to predict the impact on soil processes,
72 methods to measure the PSD have been evolving, an endeavor that is greatly complicated
73 by the extreme heterogeneity of soils, and in particular by the presence of a wide range of
74 pore sizes and morphologies. Over the years, various techniques have been proposed to
75 evaluate the PSD, based alternatively on the analysis of moisture retention curves or
76 nitrogen adsorption isotherms, or on mercury intrusion porosimetry (Echeverría *et al.*, 1999;
77 Filimonova, Hajnos *et al.*, 2006; Dexter *et al.*, 2008; Dal Ferro *et al.*, 2012). However, each
78 of these methods still suffers from a number of limitations. A common one relates to the fact

79 that the resulting pore size distribution is unavoidably influenced by the connectivity of pores,
80 between the inner portion of samples and their periphery. Furthermore, none of the available
81 techniques can detect isolated pores, which, as a result of the dynamic nature of soil
82 structure, may become reconnected over time. The analysis of N₂ adsorption method is
83 suitable only for small pores less than 0.1 μm in diameter, whereas the determination of the
84 PSD based on the moisture retention curve runs into difficulties in swelling soils, because of
85 pore drainage and shrinkage (Zong *et al.*, 2014).

86 The major technological advances in non-destructive imaging techniques that
87 occurred in the last decade, in particular the commercialization of affordable bench-top X-ray
88 Computed Tomography (X-ray CT) systems, have changed dramatically the way we look at
89 the internal geometry of soil voids (Ketcham and Carlson, 2001; Wildenschild *et al.*, 2002;
90 Kaestner *et al.*, 2008; Taina *et al.*, 2008; Wildenschild and Sheppard, 2013). Especially since
91 the development of efficient, non-operator-dependent algorithms to segment the grayscale
92 images provided by CT scanners (Sheppard *et al.*, 2004; Iassonov *et al.*, 2009; Baveye *et al.*,
93 2010; Schlüter *et al.*, 2010; Hapca *et al.*, 2013; Houston *et al.*, 2013a; Schlüter *et al.*,
94 2014), it is now possible to get a reliable perspective on how intricate and convoluted the
95 geometry of soil voids is, down to submicron scales.

96 These past few years, various algorithms have been proposed to extract PSDs from
97 3D CT images. Unlike with other soil characteristics, e.g., porosity and specific surface area,
98 for which there is a clear consensus over the estimation approach, there is no general
99 agreement, nor a clear sense of direction regarding an appropriate method for estimating
100 PSDs. Several algorithms have been proposed (Table 1), each of which has limitations in
101 terms of pore space model representation. In many cases, authors developed software to
102 address specific situations. It is unclear if these developments were driven by a lack of
103 familiarity with existing methods, by specific computational or programming language
104 constraints, or by authors seeking further improvement of existing methods. Many of these
105 available methods do however share common algorithms (Table 1), which raises the
106 question of whether generalisations can be made. A few methods have reached the stage of

107 user-friendly software that is either commercially (e.g., Avizo) or freely available (e.g.,
108 ImageJ, Quantim4). Even though these various methods often share the same theoretical
109 basis, specific requirements associated with their application in various disciplines, like
110 hydrology or ecology, have led to PSDs being reported in different ways, either as pore
111 volume and surface distribution per class size, or as body and throat population distribution
112 per class size. Conceptually, this poses no real problem, as indeed distinct formulations may
113 be more appropriate in particular cases than in others, but it has made it difficult to compare
114 the performance of the different algorithms and to determine their limitations.

115 In this general context, the objective of this study is to review existing PSD estimation
116 methods from both a theoretical and practical perspective, and to compare their performance
117 on a selection of synthetic 3D images as well as X-ray CT images of soils of different types.
118 The computer packages selected for this comparison have all been used in the past to
119 determine the PSD of soils, represent distinct types of algorithms (see details below), and are
120 all readily available. They include, respectively, the commercially licenced programs **Avizo**
121 (FEI Visualization Sciences Group) and **CTAnalyser** (Skyscan-Bruker), freely available
122 ImageJ plugins BoneJ and Skeletonize3D (Arganda-Carreras et al. 2008), used in
123 conjunction with the “Exact Signed Euclidean Distance Transform” (Borgefors, 1986), and
124 hereafter referred to as **DFS-FIJI**, the 2005 open-source release of **3DMA** (Lindquist *et al.*,
125 2000), the open-source library **Quantim4** (Vogel, 1997; Vogel and Roth, 2001), and the
126 program **DTM** developed by Monga *et al.* (2007, 2009) based on Delaunay triangulation.

127

128 **2 Theoretical approaches to PSD estimation and applications**

129 Despite the plethora of methods that have been developed, some common steps and
130 methods can be identified. The common steps consist of first identifying objects within the
131 image, then estimating a size measure per object, and finally forming a distribution from
132 these measures. In the case of a natural porous medium such as soil, the first of these steps
133 can be made difficult by the occurrence of tortuous interconnected pore clusters. Such
134 clusters are considered to be composed of pore bodies that connect with each other and

135 each such connection may be described as a pore throat (Lindquist and Venkatarangan,
136 1999). Much effort has been invested during recent decades into automatic methods for
137 identifying pore bodies and throats within digital images. All methods make use of a
138 dichotomous image consisting only of pore object versus solid background. The distance
139 transform (Borgefors, 1986) is embodied in many approaches, since the resulting Distance
140 Map (DM) image has numerous uses. It transforms a classified image (e.g., pore versus
141 solid) into a DM image whose elements are assigned a value representing their distance
142 from the nearest pore-solid interface. Local maxima of the distance transform define points
143 that can be used to extract the medial axis of objects, and also offers a means of
144 accelerating search procedures on the object space. The tools of mathematical morphology
145 (Serra, 1982) also appear within several approaches, as a means of extracting the discrete
146 skeleton (a homologue of the medial axis) as well as other transformations of pore objects.
147 The main techniques for identifying throats and bodies within segmented images include
148 **medial axis extraction, maximum inscribed balls, morphological opening, and object**
149 **separation by watersheds** (see Table 1). A brief description of these techniques is
150 presented in the following sections.

151 **2.1 Medial axis**

152 The medial axis, first proposed by Blum (1973) as an image analysis tool for object shape
153 recognition, has been intensively used for the purpose of pore space modelling. It is defined
154 as the topological skeleton running through the middle of pore channels. Several
155 approaches for medial axis extraction have been proposed, including skeletonization by
156 morphological thinning or burning algorithms, methods based on distance transform and
157 Voronoi tessellation methods.

158 **The morphological thinning** approach operates directly on the binary image,
159 resulting in a discrete image description of the pore space skeleton (Baldwin *et al.*, 1996).
160 The process is based on iterative application of morphological erosion operations, which
161 must be constrained and ordered according to a local topological structure within the image
162 (Lee *et al.*, 1994). The iterative application leads to the pore space skeleton, then a

163 skeleton distance function is defined as the Euclidean distance from each skeleton pixel to
164 the nearest solid pixel. However, despite the use of constraints, there is no guarantee of a
165 uniquely determined result, especially for pore objects that are asymmetric with respect to
166 the skeletal axis. Analogous to the thinning method is **the pore space burning algorithm**
167 (Linguist et al., 1996) which can be described as a fire that starts at the pore boundary and
168 spreads with uniform speed burning everything in its path until the different wavefronts
169 eventually meet in the middle. The set of all points where the fire directionally extinguishes
170 itself provides the skeleton of the medial axis. A size measure is given by the time at which
171 the fire reaches any unburned point, known as the burn number.

172 Another approach to medial axis extraction relies on the use of the distance
173 transform to detect **ridges (local maxima) in the distance map** image via analysis of zero-
174 crossing points in its spatial gradient (Siddiqi and Pizer, 2008). Once the location of the
175 medial axis points has been determined, accurate geometric description (i.e. including
176 surface orientation) of the medial axis can be obtained using the structure tensor (Heyden
177 and Kahl, 2011). This is a covariance matrix formed from weighted combinations of gradient
178 vectors in the local neighbourhood of a point. The eigensystem of this covariance matrix
179 reveals local anisotropy in object structure and hence can be used to infer dimensionality
180 (point, line or plane). A disadvantage of this approach is the computational cost: A large
181 number of covariance matrices must be constructed and their eigensystems determined.

182 **Voronoi tessellation** has also been proposed for medial axis extraction. It consists
183 of partitioning the pore space into 3D Voronoi regions based on seed points placed on the
184 boundary of the pore objects (Delerue *et al.*, 1999; Delerue and Perrier, 2002). The medial
185 axis can then be extracted from the the subset of the Voronoi facets located inside the pore
186 surface and further filtering according to some angle criteria. A size measure for these
187 features can be determined from a distance transform image (computed separately) or by
188 explicit search. If material exhibits a resolved granular structure with well defined pore
189 objects, this approach provides a good approximation of the medial axis. In general,
190 however, the method is highly unstable with respect to small details of pore shapes.

191 Therefore, for pore objects that are irregular and complex in shape, an alternative is to use
192 Delaunay triangulation to decompose the boundary of pore objects into 3D surface
193 elements (Monga et al, 2007, 2009). Voronoi regions are then produced from these surface
194 elements and filtering is applied as before to approximate the medial axis. For a precise
195 description of network structure, the decomposition into surface elements may have to be
196 very detailed which leads to extreme computational cost. In practice a balance between
197 accuracy and smoothness is achieved by locally adapting the surface tessellation.

198

199 **2.2 Maximum inscribed balls (MIB)**

200 This technique finds the largest inscribed spheres centred on each voxel of the pore space
201 that just touches the pore surface. Those that are fully overlapped by larger spheres
202 (engulfed) are removed; the remaining spheres are called maximal balls and cover fully the
203 pore space. Within the pore-ball description, balls that touch or overlap are considered linked
204 to one another by pore channels, hence a graph description consisting of nodes (balls
205 representing pore space) and edges (the channels linking pore space) may be extracted
206 (Silin and Patzek, 2006). Finding the minimal set of maximum-sized balls that accurately
207 describe pore space, requires a search procedure to locate all engulfed balls and then
208 eliminate them from the pore-ball description. This is straightforward in the case of a “simply
209 engulfed” ball but challenging in the case of “compound engulfment”. The combinatoric
210 nature of this search problem means that the algorithm employed must be considered
211 carefully in relation to problem size and computational capacity. As a result some
212 implementations of the MIB procedure make use of medial axis function as a support to fit
213 the inscribed spheres, reducing in this way considerably the search space.

214

215 **2.3 Morphological opening**

216 This algorithm iterates over increasing level thresholds on the distance map of the pore
217 space, constructing both an "opening map" image and also a mask image that guides
218 subsequent iterative construction (Vogel, 1997). Within each iteration, a morphological

219 structuring element (a ball of radius indicated by the current distance threshold) is applied at
220 locations on the boundary of the solid background as dictated by the mask image. The
221 opening map records the distance threshold at which each image element has been so
222 "opened", while the mask image helps eliminate redundant operations. Although the
223 distance map may use the Euclidean distance metric, the reliance on morphological
224 operations means that it is impractical to generate an opening map of continuous Euclidean
225 distance measure. Only integer-valued distances are recorded, hence the opening map
226 contains a subset of the Euclidean measure, considered in the present work to be a
227 "morphological distance measure".

228

229 **2.4 Object separation method**

230 This technique makes use of a distance transform of the binary image to create a distance
231 map to which a watershed transformation is applied to separate the pore space into pore
232 objects (Rabbani *et al.*, 2014). This is achieved by identifying watershed basins around each
233 local maximum of the distance transform, resulting in one pore object associated with every
234 local maximum. When pores have a rough surface, application of this technique can break
235 the pore space into many small objects due to additional local maxima near the surface. A
236 main limitation of this partitioning method is the use of spherical structuring elements when
237 identifying watershed basins, which might not cope very well when subject to tortuous
238 interconnected pore clusters.

239

240 **3 Materials and Methods**

241 Performance of existing PSD software was evaluated on a selection of X-ray CT soil images
242 as well as 3D synthetic images that were constructed based on a simple 3D ball pore
243 geometry at different porosity levels. Comparison was possible among the methods
244 providing the same type of PSD output, either in the form of pore volume fraction per size
245 interval (for BoneJ, CTAnalyser, Quantim4 and Avizo), or object population fraction per size
246 interval (for 3DMA, DFS-FIJI and Avizo). Avizo was the only software in the study that would

247 provide both types of outputs. Additionally, for the synthetic images it was possible to
248 compare the methods against the exact analytical solution.

249

250 **3.1 Image data**

251 **Soil images**

252 Undisturbed soil samples were obtained from the top 5 cm of Mid Pilmore at the James
253 Hutton Institute (JHI, Dundee, UK) under three tillage regimes (no tillage, minimum tillage
254 and ploughed), as previously described by (Sun *et al.*, 2010; Pérez-Reche *et al.*, 2012;
255 Hapca *et al.*, 2013; Houston, *et al.*, 2013b; Juarez *et al.*, 2013). This gave four soil
256 treatments with three replicates per treatment, yielding a total of 12 samples.

257 Soil images were obtained using a HMX225 X-ray micro-tomography system (NIKON
258 Metrology, UK). The undisturbed samples were scanned at 150 kV and 50 μ A using a 2 mm
259 aluminium filter to obtain 1200 angular projections with 4 exposures per frame. A
260 molybdenum target was used. The repacked samples were scanned at 125 kV and 131 μ A
261 using a 0.5 mm aluminium filter and 3010 projections. All radiographs were reconstructed
262 into a 3D volume using CT-Pro v.2.0 (NIKON Metrology, UK). For each sample a 512^3
263 voxels region of interest at the centre of the sample volume was selected and reconstructed
264 at 50 μ m resolution. Reconstructed images were mapped from 32-bit floating point to 8-bit
265 unsigned using the outlier rejection method (Houston, *et al.*, 2013b). Segmentation was
266 achieved using Adaptive Window Indicator Kriging (Houston, *et al.*, 2013a) incorporating
267 hysteresis threshold determination as described in Schlüter *et al.* (2010). Standard
268 morphological measures of the pore space including porosity, pore surface area, and
269 connectivity were calculated for each of the soil samples and used as an initial soil treatment
270 comparison. The porosity was calculated as the image volume fraction occupied by the pore
271 space, the pore surface area was computed for each segmented image according to the
272 prescription in Ohser and Mücklich (2000). The surface area is a dimensionless parameter
273 being calculated relative to the outer surface area of the cube in order to enable for

274 comparisons of different volumes. The connectivity was estimated as the volume fraction of
275 pore space that connects with the surface of the image volume (Houston et al., 2013b).

276

277 ***Synthetic images***

278 Synthetic images were constructed algorithmically by applying a constrained boolean model
279 of 3D balls to create the pore space. The objective of this approach is to obtain images
280 containing clusters of pore bodies, the surface of each cluster being a set of truncated
281 spheres that inter-connect by circular pore throats. In addition to these clusters, a number of
282 non-intersecting spherical pore bodies are also typically present within such images. The
283 choice of spherical pore bodies can be motivated by the fact that it leads to relatively simple
284 design and implementation of the synthesis algorithm, which allows one to formulate
285 appropriate constraints and to determine analytic measures. Specifically, by using only
286 spheres, it allows every pore body to be easily identified and clearly discriminated from all
287 others, at the same time it ensures that every throat aperture is significantly smaller than the
288 bodies it connects.

289 Once the network structure of pore bodies associated with the synthetic image is
290 available (Figure 1), the procedure is to inspect individual clusters and to delete the smaller
291 ball whenever the overlap between a pair of balls does not meet prescribed criteria. The
292 criteria are selected so as to ensure first that a circular “throat” of intersection is
293 unambiguously defined in every case, and second that each pore body is enclosed by a
294 spherical surface that can be clearly discriminated from that of all other bodies. As well as
295 limiting the degree of overlap (in terms of volume) between any pair of balls, it is important to
296 detect any overlap of throat intersection between three or more balls. This latter condition
297 implies combinatorial processing of the ball descriptions, i.e., each ball needs to be checked
298 against all others in all possible combinations. In practice however, it suffices to detect and
299 rectify triple intersections because quadruple or larger intersections may be decomposed
300 into conjunctions of triple intersections. Given that for every case of multiple intersections, all
301 balls except the two largest ones are deleted, there is no dependency upon the order in

302 which triple intersections are detected. As a result a searching algorithm of $O(n^3)$ order was
303 applied as a simple “brute force” approach to deal with the multiple intersections problem.
304 Additionally, individual spherical bodies are permitted to intersect, subject to a number of
305 constraints, so as to produce more complex pore networks. The purpose of the constraints is
306 to ensure a well-defined circular throat aperture between each intersecting body pair. This
307 means ensuring that the distance between the centre points of overlapping spheres is
308 neither too large nor too small, and also that each circular aperture is distinct from all others.
309 The criteria presented above were used to generate three synthetic images with parameters
310 chosen so that to produce different porosity levels (0.17, 0.24 and 0.29, respectively). For
311 the first two images, additional constraints were used to ensure that individual bodies were
312 fully contained within the image, without intersecting the image boundaries. An exception
313 was made in the case of the third sample (of 0.29 porosity), for which a number of spherical
314 bodies were permitted to touch (without being truncated) the upper and lower surfaces of the
315 image, creating a vertically percolating pore network. For each synthetic image, an exact
316 analytical measure of the pore size distribution was defined by labelling each ball with the
317 corresponding diameter and calculating the relative frequency of balls per size diameter to
318 derive a measure of object population per size interval or, alternatively, by calculating the
319 pore volume occupied by balls of same diameter to derive a measure of pore volume fraction
320 per size interval. The exact analytical solution was further compared with those obtained by
321 the various algorithms.

322

323 ***3.2 Image preparation and application of specific PSD analysis methods***

324 All the PSD methods make use of a binary image consisting only of pore objects versus solid
325 background, converted as necessary to compatible file formats such as TIFF, BMP or RAW
326 format images. It was in some cases necessary to designate object versus background
327 image elements. The exceptions to this include Quantim4 and 3DMA, both of which implicitly
328 identify zero-valued elements as being pore (displayed black). Another exception is ImageJ
329 plugins, which typically identify objects as consisting of the 8-bit element value 255 (usually

330 displayed white) and so require black-pore-object images to be inverted. The specific
331 sequence of operations required for each analysis method together with a brief description of
332 the underlying theoretical approach is given in the supplementary material.

333

334 **3.3 Statistical evaluation of the PSD analysis results provided by the different** 335 **methods**

336 Performance evaluation and comparison of the PSD methods presented above was
337 conducted on the synthetic images and the soil image data.

338 Statistical analysis of the PSD results was conducted by fitting a two-parameter
339 gamma distribution model to the PSDs provided by the different methods for each of the
340 fifteen image samples. The gamma distribution was chosen on the basis that it is a positive
341 distribution, which, depending on the values of the two parameters (shape and scale
342 parameters), can be very flexible in covering a variety of shapes ranging from positively
343 skewed to symmetric. As a result the gamma distribution was a good model candidate to fit
344 the different shapes of the PSDs produced by the different methods and the different soil
345 types or synthetic images. The Non-Linear Mixed-Effect procedure in R (*nlme* package in R
346 v.3.1.1) was used to fit the gamma distribution to the data and to investigate significant
347 difference in the PSD model parameters (for both shape and scale simultaneously)
348 estimated for the different methods. Method comparison was conducted first on all the soil
349 images (twelve samples). For methods performance comparison on the soil data, methods
350 and soil treatments (with four levels no tillage, minimum tillage, ploughed and sieved).
351 were introduced in the model as fixed factors and the soil samples as random factors. To
352 assess the consistency of the methods throughout the soil treatments an interaction terms
353 between methods and treatments were also investigated. A second analysis was also
354 conducted on each of the soil treatment samples (three replicates) separately and on the
355 synthetic images (three replicates). In this analysis, methods were introduced into the model
356 as fixed factors and samples as both fixed and random factor. To assess the consistency of
357 the methods throughout the different samples, interaction effects between methods and

358 samples were also investigated. The methods comparison analysis was performed
359 separately, first for the pore volume fraction based PSD methods and then for the object
360 population fraction based methods.

361

362 **4 Results**

363 ***4.1 PSD of the synthetic image data***

364 The distributions of pore volume fraction per size interval estimated by Bone J, CTAnalyser,
365 Quantim4, Avizo and DTM are in good agreement with the analytical solutions. The
366 parameters of the gamma distribution fitted to the PSDs estimated by these five methods are
367 not significantly different from the analytical solution (p -values >0.81). As illustrated in Figure
368 2, these results are consistent for all three synthetic samples. Avizo and DTM seem to
369 generate some fictitious small diameter results, however. The distributions produced by
370 Avizo and DTM also exhibit some slight irregularities compared to the other three methods.
371 In the case of Avizo, this may be linked to problems evident within the separated object
372 maps, where the separating surfaces in some cases seem excessive in number, giving rise
373 to fragmentary objects.

374 Estimation of PSD by 3DMA, DFS and Avizo in terms of object population fraction
375 per size interval, shows significant interaction effects between the different methods and the
376 three synthetic image samples (p -values <0.001), indicating that these methods are not
377 stable in their estimation when subjected to a range of pore space morphologies. Compared
378 to the analytical solution (Figure 3), Avizo has a tendency to overestimate the pore
379 population fraction of small class size in the sample with small porosity (A1 -17% porosity).
380 The DFS method in general, overestimates the population fraction of small pores, whereas
381 the 3DMA method overestimated the population fraction of large class size pores.

382

383 ***4.2 PSD of the soil image data***

384 Standard morphological measures of the four treatments, including porosity, pore surface
385 area and connectivity are presented in Table 2. Soil porosity and connectivity were not

386 significantly different among no-tillage, medium-tillage and ploughed treatments (p-
387 values>0.05) whereas the sieved soils had a porosity and connectivity significantly lower
388 than the other three treatments (p-values<0.001). In terms of pore surface area only the
389 ploughed soils appear to be significantly different from sieved soils (p-value<0.001) and
390 medium-tillage soils (p-value<0.05), all the other pairwise comparisons being not significant
391 (p-value>0.05), possibly reflecting a relatively large within-treatment variability (Table 2).

392

393 The PSD analysis of soil data, expressed as *pore volume per class size*, revealed that
394 BoneJ, CTAnalyser and Quantim4 are in close agreement with each other but differ
395 significantly from both AVIZO and DTM (in the scale parameter, p-value<0.001 and p-
396 value=0.002 respectively). Examination of the map images for MIB methods (BoneJ,
397 CTAnalyser, Quantim4, and DTM) reveals that many partially filling ball objects are created
398 where pores have a complex shape, a feature that is widespread in the case of soil pores.
399 This feature is very clear in 3D images, but is unfortunately hard to convey adequately in 2D
400 images. Discrepancies in our perception of the connectivity and geometry of the pore space
401 based on 2D and 3D images are well known and unavoidable (Hapca et al., 2011, 2015).
402 Detailed analyses should therefore be based on 3D images. Nevertheless, the cross-section
403 in Figure 4 illustrates well the fact that in some of the wide, complex-shaped pores, instead
404 of having large balls of the relevant diameter, one often finds several smaller balls,
405 occupying less volume. As a result, the MIB based methods produced larger estimates of
406 the pore volume occupied by small pores with less volume being occupied by large pores,
407 compared to Avizo (Figure 5). This tendency gets even more noticeable in the case of PSDs
408 calculated by DTM, which is much skewed at the lower end indicating that most of the large
409 pores get fragmented into very small pores. Comparison of the PSDs for the different soil
410 treatments based on DTM indicates that no-tillage and minimum-tillage treatments were not
411 significantly different in terms of PSD shape and scale parameters (p-values>0.11), whereas
412 all other pairwise treatment differences were significant (p-value<0.05). In turn, based on
413 Avizo only, the sieved and no-tillage treatment appears to be significantly different in terms

414 of PSD scale parameter (p -value=0.046), all other pairwise treatment differences being not
415 significant (p -values>0.08). BoneJ, CTAnalyser and Quantim4 methods were consistent with
416 each other showing that the no-tillage and minimum-tillage treatments were not significantly
417 different in terms of PSD shape and scale parameters (p -values>0.28). The same thing
418 happens with the ploughed and sieved treatments (p -values>0.22). As illustrated in Figure 5,
419 the PSD of the no-tillage and minimum-tillage treatments share similar profiles with more
420 pores of larger size as compared to the ploughed and sieved treatments. The above analysis
421 shows that different methods obtain different estimates for PSD, and assessments of
422 treatment effects are affected by the method chosen.

423 Comparison of soil PSDs provided by 3DMA, DFS and Avizo in terms of *object*
424 *population fraction per class size* showed significant differences among methods for all four
425 soil treatments (p -values<0.05). In addition, PSD estimation by the three methods was
426 inconsistently different for the different soil treatments, the fitted gamma model indicating
427 significant interaction effects between methods and treatments (p -values<0.05). As
428 illustrated in Figure 6, for the sieved soil and the no-tillage treatment, there is a relatively
429 good visual agreement in the PSD estimation in particular for classes of larger size, however
430 for the ploughed treatment there is an obvious discrepancy between the methods, with Avizo
431 providing larger frequency estimates of large class size pores as compared to the other two
432 methods (p -values<0.05). As for the minimum-tillage treatment, all three methods appear to
433 disagree in their PSD estimation (p -values<0.05). In particular, the DFS method this time
434 appears to overestimate the frequency of large class size pores as compared to Avizo and
435 3DMA methods. Comparison of the different soil treatments based on Avizo indicated that
436 the sieved soils were significantly different from all the other treatments in terms of PSD, with
437 all the other pairwise treatment comparison not being significant (p -values>0.10). In turn, the
438 DFS method identified significant differences in terms of PSD between the ploughed soil and
439 the other treatments, and between the sieved treatment and the no-tillage treatment (p -
440 values<0.05), all other pairwise treatment comparison not being significant (p -values>0.08).
441 Finally, the 3DMA method identified all soil treatments as being significantly different in

442 terms of PSD. The lack of agreement between these methods suggest that object population
443 fraction based PSD methods such as 3DMA, DFS or Avizo are not necessarily suited for soil
444 data, in particular for the purpose of soil treatment comparison.

445

446 **5 Discussion and conclusions**

447 The Pore Size Distribution (PSD) has been widely used as a means of characterising the
448 physical structure of geomaterials including soils, since at least the mid-20th century, with
449 links to both fluid transport properties and the availability of ecological habitat. However, for
450 soils, which are very heterogeneous in their physical structure due to a wide range of pore
451 sizes and morphologies, estimation of the PSD is particularly challenging. Despite
452 significant work on the development of both traditional invasive techniques and non-
453 destructive 3D image analysis methods, there is still no consensus on what method should
454 be used. In this context, the purpose of our work was to present a theoretical review of
455 underlying methodologies and to compare available methods for application in soil science
456 through a statistical framework.

457 The statistical framework developed in this study for PSD method comparison is
458 based on a gamma distribution model fitted to the PSDs estimated by the different methods
459 for the different soil types. Then, a nonlinear mixed-effect procedure was considered in order
460 to statistically compare the estimated parameters of the gamma distribution model for the
461 different cases. To our knowledge, this is the first time a robust statistical method is
462 developed and used for the purpose of PSD comparison. In the last few years, a number of
463 authors have instead proceeded to a visual comparison of PSDs (e.g., Al-Raoush *et al.*,
464 2003, Al-Raoush and Wilson 2005; Dong *et al.*, 2008; Ngom *et al.*, 2011). In principle, these
465 two approaches could be viewed as complementary. Our perspective, nevertheless, is that ,
466 as with the methods used to threshold CT images (Baveye *et al.*, 2010), an approach that is
467 objective, *i.e.*, does not rely on operator judgment, is likely to lead to more reliable
468 conclusions.

469 A total of seven methods were considered for assessment in this study, which
470 include the commercially licenced Avizo and CTAnalyser, freely available plugins BoneJ and
471 Skeletonize3D (called here DFS-FIJI) for ImageJ, the 2005 open-source release of 3DMA,
472 as well as the open source libraries Quantim4 and DTM. It was found that all methods
473 presenting the PSD as pore volume per class size (this includes Avizo, CTAnalyser, BoneJ,
474 Quantim4 and DTM) were in good agreement with the analytical solution when tested on the
475 synthetic images. Avizo makes use of spherical structuring elements when identifying
476 watershed basins, while the other four methods share an MIB-based approach to PSD
477 calculation, which explains the good agreement with the analytical solution on the synthetic
478 samples. In turn, a great discrepancy was found between the analytical solution and the
479 methods for which PSD is calculated as object population fraction per class size, in particular
480 for 3DMA and DFS-FIJI. Differences in method estimation appeared to get even wider in the
481 case of soil images, with only CTAnalyser, BoneJ and Quantim4 providing consistently
482 similar distributions for the different soil types, the rest of the methods being all different.
483 These findings are in agreement with some previous studies (Al-Raoush *et al.*, 2003, Al-
484 Raoush and Wilson, 2005; Dong *et al.*, 2008; Ngom *et al.*, 2011), which have also reported
485 differences among the PSD estimation methods tested. In the study by Al-Raoush *et al.*
486 (2003), 3D images of synthetic structures of spheres regularly and randomly packed were
487 used to compare the performance of a medial axis approach for pore network extraction
488 against a method based on modified Delaunay tessellation. The two methods provided
489 similar PSD results when tested on synthetic regular packing, but great discrepancies were
490 found when the methods were applied to randomly packed spheres. In a different study,
491 Dong *et al.* (2008) compared four methods, medial axis (Lindquist *et al.*, 1996), maximal ball
492 (Sillin and Patzek, 2006), velocity based (Øren *et al.*, 2006) and grain recognition based
493 algorithm (Øren and Bakke, 2003), on 3D rock microstructure images of both sandstone and
494 carbonate obtained from process based reconstructions and X-ray micro-tomography. Again
495 it was reported that depending on the type of structure and type of images, there is a
496 difference in the level of agreement among PSD estimates provided by the four methods. In

497 particular very little agreement was found for those images presenting pores of low sphericity
498 angular shapes. In a more recent study by Ngom *et al.* (2011), a Delaunay Triangulation
499 Method (DTM) for PSD estimation (Monga *et al.*, 2007, 2008) was compared against the
500 3DMA method (Linguist *et al.*, 2000) based on two soil samples from two different
501 treatments, a ploughed soil and a grassland soil. It was reported that for both sample images
502 the DTM method tended to fragment the pore space into small pores resulting in PSD with a
503 higher pick at small class sizes as compared to the 3DMA method, which presented lower
504 pick at small class sizes but longer tails for large class sizes.

505 In the current study, the DTM method was also compared against several other
506 methods for PSD estimations. It was found that while on synthetic images the DTM was in
507 good agreement with the other methods and with the analytical solution, when tested on the
508 soil images the PSD distribution generated by the DTM method was very skewed to the
509 lower end due to many fragmentary pore objects being created for soil pores with complex
510 shape. In turn the watershed-based Aviso method appears to separate the pore space into
511 larger objects as compared to the MIB-based method resulting in low peak, longer-tailed
512 PSDs. However, despite these clear differences in overall performance on soil images
513 (Figure 5), a characteristics of the volume per class size PSD estimation methods was the
514 consistency in the PSD profiles produced by these methods independent of the soil type,
515 suggesting only mild interaction effects between the methods and the soil type on the PSD
516 estimation. This further indicates that each of these PSD estimation methods can be reliably
517 used for the purpose of soil type assessment and comparison.

518 The second type of methods, based on object population fraction per class size, was
519 less consistent in terms of PSD estimation when applied to both synthetic images and soil
520 image data. In the case of the synthetic images, the 3DMA and DFS estimations for PSD
521 were different from the analytical solution for all three synthetic images. The DFS method
522 appeared to fragment the pore space into smaller pores resulting in PSDs being more
523 skewed towards the lower end as compared to the analytical solution, and this performance
524 was consistent for all three synthetic images. In turn the 3DMA method produced completely

525 different distribution profiles for the three synthetic images with the low porosity image
526 (sample 1), having a longer tailed distribution compared to the analytical solution while for
527 the high porosity sample (sample 3) the distribution had a higher pick at the lower end as
528 compared to the analytical solution (Figure 3). The Avizo method was in agreement with the
529 analytical solution for the high porosity samples, but failed to perform well on the low
530 porosity sample (sample 1), identifying more pore objects of larger size than in reality. An
531 explanation to this can be that for the low porosity sample there is less degree of overlap
532 between the 3D ball objects, and therefore for the overlapping balls the shape of the objects
533 are not too complex to be separated by the watersheds (in particular when a small ball
534 overlaps with a large ball), and so this is kept as one big pore object during the PSD
535 estimation. When applied to soil images, the level of agreement between the three methods
536 depended on the soil type; for the no-tillage and sieved soil all three methods provided very
537 similar PSDs, whereas for the medium tillage and ploughed soils the estimation in the PSD
538 by the different methods was very different. This inconsistency in method performance when
539 applied to different soil treatments was statistically confirmed by the significant interaction
540 effects between methods and the soil types when the gamma model was fitted to the data
541 (Figure 6). This further indicates that the PSD methods based on object population fraction
542 per class size are less reliable to be used for the purpose of soil treatment assessment and
543 comparison.

544 In general, the lack of agreement among the PSD estimation methods can be
545 attributed to the way each of these methods handle tortuous interconnected pore clusters or
546 rough surface pores, which can lead in some cases to many fragmentary small objects being
547 created along the pore surface. The volume contribution of these small objects is still
548 negligible and therefore volume-based PSD methods are less affected by these artefacts,
549 whereas if a large amount of small pore objects is created, this can have a high impact on
550 the shape of PSDs reporting the relative frequency of objects per class size.

551 Whereas this study presents an up-to-date theoretical and practical assessment of
552 existing methods for PSD estimation from 3D porous media images, of main interest is the

553 performance of PSD methods on 3D soil images, which pose additional challenges due to
554 the heterogeneous nature of the inner pore structure. Based upon the current analysis, we
555 recommend that PSD be presented as a pore volume per size class, which for the methods
556 tested gave the greatest consistency and confidence that the methods can be used for
557 relative comparisons of samples. Of the methods tested, Bone J, CTAnalyser, Quantim4,
558 Avizo and DTM were in good agreement with the analytical solutions for pore volume per
559 size class. For soil however, only the methods based on MIB (Bone J, CTAnalyser,
560 Quantim4) produced consistent results. We also found that methods based on object
561 population fraction per class size produced unstable results for both the synthetic samples
562 but in particular for the more complex soil samples. We therefore recommend that these
563 methods be avoided till improved further.

564

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Tables

Table 1. Examples of methods for pore size distribution estimation, the main algorithm they are based upon and the main measures they produce as output to present the pore sizes in a porous medium. The reported studies had a **main** element of method development or application and are all based on information from 2D thin sections or 3D data sets obtained with X-ray CT. See text for description of main algorithms and methods.

Publication	Software	Method
<i>Baldwin et al. 1996</i>	Autors - development	Medial axis by morphological thinning
<i>Lindquist et al., 1996, 2000</i>	3DMA - development	Medial axis by morphological thinning/burning of pore space
<i>Vogel 1997</i>	Quantim4 - development	Morphological opening by erosion and dilation using an incremental spherical structuring elements
<i>Vogel and Roth, 1998, 2001</i>	Quantim4 - application	Morphological opening (Vogel 1997)
<i>Lindquist & Venkatarangan, 1999</i>	3DMA - application	Medial axis by morphological thinning (Lindquist <i>et al.</i> 1996)
<i>Delerue et al., 1999</i>	Authors - development	Medial axis by Voronoi tessellation, maximum inscribed balls
<i>Thovert et al., 2001</i>	Authors - development	Maximum inscribed balls
<i>Lindquist, 2002</i>	3DMA - development	Medial axis by morphological thinning
<i>Delerue and Perrier, 2002</i>	DXView - development	Medial axis by Voronoi tessellation, maximum inscribed balls
<i>Pierret et al., 2002</i>	Authors - development	Morphological opening (using a 32 face "sphere" structuring element)
<i>Ams, 2004</i>	Author - application	Maximum inscribed balls (Thovert <i>et al.</i> 2001)
<i>Al-Raoush and Wilson, 2005</i>	3DMA - application	Medial axis by morphological thinning (Lindquist <i>et al.</i> 1996)
<i>Silin and Patzek, 2006</i>	Authors - development	Maximum inscribed ball
<i>Prodanovic et al., 2006</i>	3DMA - development	Medial axis by morphological thinning
<i>Al-Kharusi and Blunt, 2007</i>	Authors - development	Maximum inscribed balls
<i>Jiang et al., 2007</i>	Authors - development	Medial axis by morphological thinning prioritized by Euclidean distance
<i>Monga et al., 2007, 2009</i>	DTM - development	Delaunay triangulation and maximum inscribed balls
<i>Peth et al., 2008</i>	3DMA - application	Medial axis by morphological thinning (Lindquist <i>et al.</i> , 2000)
<i>Dong and Blunt, 2009</i>	Authors - development	Maximum inscribed ball
<i>Talabi et al., 2009</i>	Authors - application	Maximum inscribed balls (Al-Kharusi and Blunt, 2007).
<i>Doube et al., 2010</i>	BoneJ (ImageJ) - development	Maximum inscribed ball, medial axis by finding ridges on an Euclidean distance map
<i>Luo et al., 2010</i>	Avizo5 - application	Object separation, 3D skeletonization.
<i>Kravchenko et al. 2011b</i>	3DMA - application	Medial axis by morphological thinning (Lindquist <i>et al.</i> , 2000)
<i>Ngom et al., 2011</i>	DTM and 3DMA - application	Delaunay triangulation (Monga <i>et al.</i> , 2007, 2009), medial axis (Lindquist <i>et al.</i> 1996)
<i>Vaz et al., 2011</i>	Authors - application	Morphological opening (Vogel and Roth, 1998; Pierret <i>et al.</i> , 2002)
<i>Beckingham et al, 2013</i>	3DMA - application	Medial axis by morphological thinning (Lindquist <i>et al.</i> , 1996, 2000)
<i>Wang et al., 2013</i>	3DMA - application	Medial axis by morphological thinning (Lindquist <i>et al.</i> , 2000)
<i>Rabbani et al., 2014</i>	Authors - development	Object separation by distance and watershed transform
<i>Naveed et al., 2014b</i>	BoneJ (ImageJ) - application	Maximum inscribed balls (Doube <i>et al.</i> , 2010)
<i>Munoz-Ortega et al. 2015</i>	Quantim4 - application	Morphological opening (Vogel 1997; Vogel <i>et al.</i> 2010)
<i>Armstrong et al., 2015</i>	3DMA - application	Medial axis by morphological thinning (Lindquist 2002; Prodanović <i>et al.</i> 2006).

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Table 2. Summary of soil morphological measures of the four soil treatment showing porosity, pore surface area, and pore connectivity sample mean \pm SE (n=3).

Morphological soil properties	No tillage	Medium tillage	Ploughed	Sieved
Porosity	0.108 \pm 0.012	0.118 \pm 0.018	0.144 \pm 0.021	0.044 \pm 0.002
Surface area	17.039 \pm 1.081	14.619 \pm 2.464	25.183 \pm 5.224	9.675 \pm 0.485
Connectivity	0.764 \pm 0.063	0.909 \pm 0.020	0.880 \pm 0.059	0.299 \pm 0.043

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813 **Legend to figures**

814

815 **Figure 1.** Graphical renderings of a synthetic sample (corresponding to 17% porosity)
816 illustrating from left to right (a) network formed by intersecting balls, (b) isolated (dark) and
817 intersecting (light) balls, and (c) colour labelling of cluster image elements based on the 6-
818 connected neighbourhood. In the latter image, only the largest 250 clusters are assigned a
819 distinct colour, the remainder are shown in transparent grey.

820

821 **Figure 2.** Distribution of pore volume fraction per size interval for the synthetic images and
822 the corresponding Gamma distribution fit; comparison of PSD methods (BoneJ, CTAnaliser,
823 Quantim 4, DTM and Avizo) against the analytical solution.

824

825 **Figure 3.** Distribution of pore object population fraction per size interval for the synthetic
826 images and the corresponding Gamma distribution fit; comparison of PSD methods (Avizo,
827 DFS-FIJI, 3DMA) against analytical solution.

828

829 **Figure 4.** (a) Illustrative cross-sectional thresholded image through one of the soil samples
830 (sample M1-1), with black pixels representing the solid phase and white pixels the pore space.
831 (b) Image of the same cross-section with the pore space approximated with balls, using DTM.
832 At the top left and at the bottom of this image, there is evidence of partial filling of pores due
833 to edge effects, which can be eliminated by selecting a smaller image after approximation by
834 balls. Throughout the image, pores with complex geometries tend to be partially filled by a
835 combination of small and slightly large balls.

836

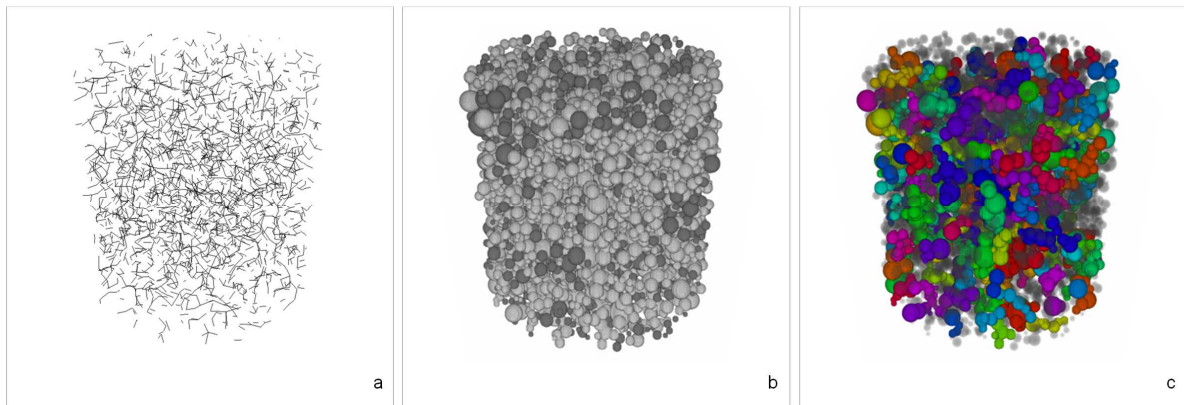
837 **Figure 5.** Distribution of pore volume fraction per size interval on a selection of soil images,
838 as calculated by BoneJ, CTAnalyser and Quantim 4, DTM and Avizo, and the corresponding
839 Gamma distribution fit. A single Gamma distribution was fitted to BoneJ, CTAnalyser and
840 Quantim4 as these three methods were found not significantly different.

841

842 **Figure 6.** Distribution of pore object population fraction per size interval on a selection of soil
843 images, as calculated by Avizo, DFS-FIJI and 3DMA, and the corresponding Gamma
844 distribution fit.

845

846 Figure 1

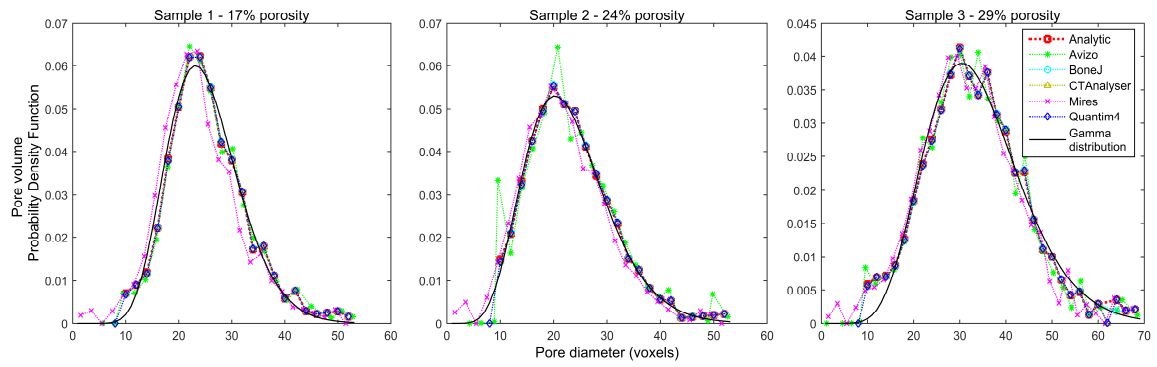


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849 Figure 2

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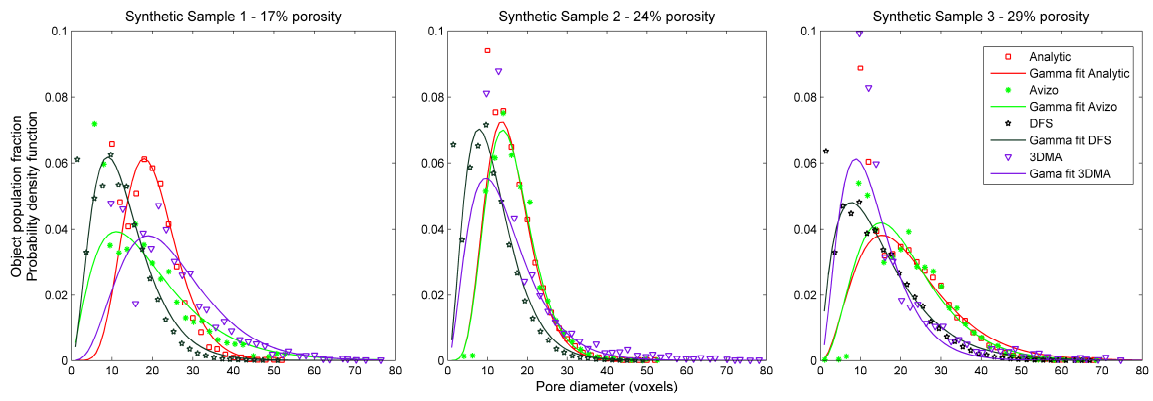


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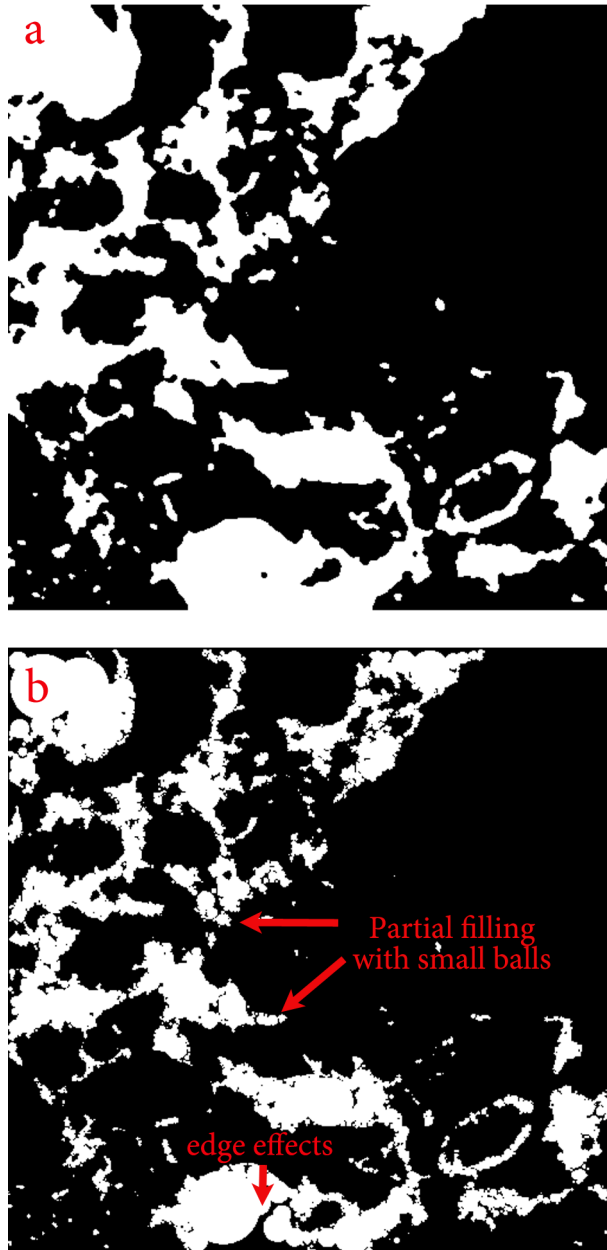
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854 Figure 3
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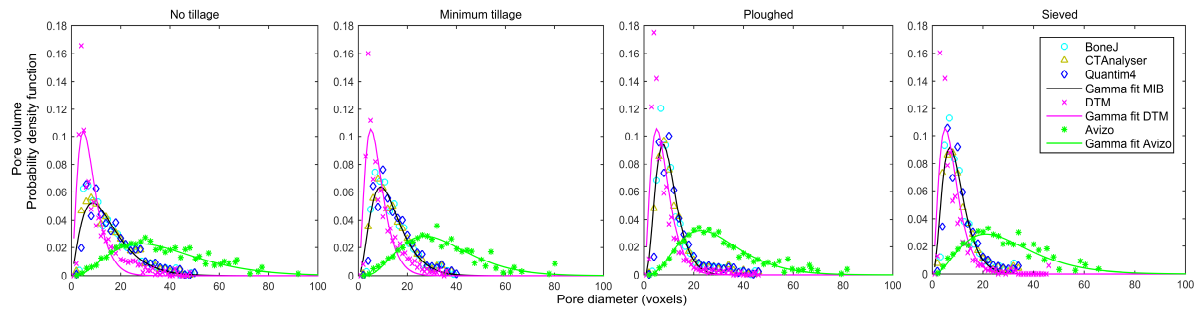
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862 Figure 4
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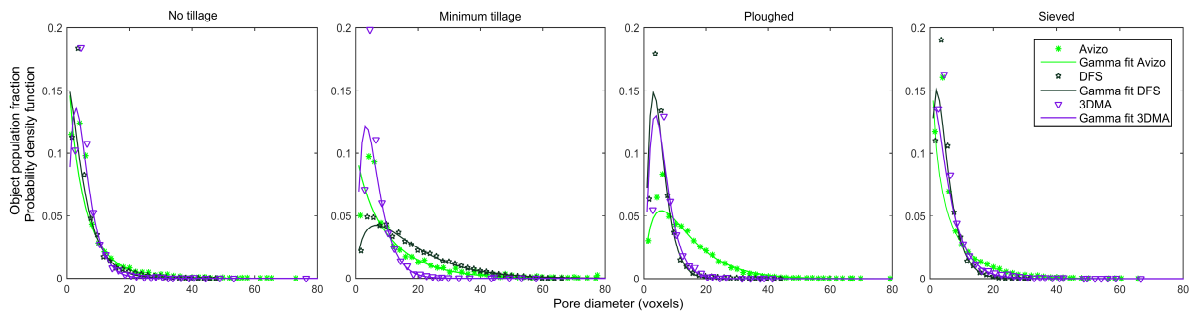
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876 **Supplementary material**

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880 **Description of the computer packages used for PSD estimation**

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882 ***Avizo Fire***

883 Avizo (version 7.1) is commercial software consisting of a base application (providing the
884 ability to process and visualise data in many formats) plus a range of optional software
885 modules that extend the basic functionality (FEI Visualization Sciences Group). The “Fire”
886 extension package provides a selection of such extensions appropriate for materials science
887 and these were used within the present study.

888 The ‘separate objects’ feature of Avizo Fire, which operates by constructing watersheds
889 within the background phase of the image, was used to divide pore objects into size classes
890 (Avizo documentation). These watersheds are allowed to project through objects in the form
891 of surfaces explicitly represented using image elements, which may result in some slight
892 distortion of measures (such as volume) on objects separated in this manner.

893 The effect of watershed separation applied to soil pores is that larger and more
894 tortuous pore clusters are partitioned into a number of smaller objects. This tends to reduce
895 the incidence of object concavity within the image, producing a population of more convex
896 objects. Each object can then be individually labelled (using region-growing on a specified
897 local neighbourhood) and finally measured using specific functions built into Avizo. For the
898 present study, labelling was carried out using the 6-connected neighbourhood and the built-
899 in measure functions “Width3d”, “EqDiameter”, “Unweighted” and “Volume3d” were collected
900 per object. The “Width3d” measure is an estimate of the Feret diameter (Merkus, 2009) and
901 “EqDiameter” is the diameter of an equivalent sphere (i.e. one whose volume equals that of
902 the object). Feret diameter was estimated using 30 samples which is the default value
903 suggested by the software. The “Unweighted” function provided the number of objects per
904 object diameter while the “Volume3d” measure provides the volume estimated by point
905 counting.

906

907 **CTAnalyser**

908 CTAnalyser (version 1.13) is provided commercially along with X-ray CT equipment as part
909 of an overall imaging solution (Skyscan-Bruker microCT). This software implements a size
910 measure entitled “structure thickness” (commonly known as “trabecular thickness” when
911 bone is analysed) by a method based on fitting maximal balls within the object. This is
912 achieved via analysis of a distance map but the metric used to form the distance map is not
913 specified in the documentation, nor is the resulting map exported. The software does
914 however permit the final size map image to be saved to disk. The elements of the size map
915 image are 8bit indices denoting the size category of the covering ball, the index zero denotes
916 background phase elements while the smallest objects are denoted by an index of one:
917 objects belonging to larger size categories are assigned indices in ascending order. An
918 accompanying report text file allows each index value to be related to a size measure and
919 also gives the volume (estimated by point counting) for each size class. After designating the
920 objects of interest using a thresholding operation, the remaining processing is fully
921 automatic, i.e. no user specified parameters are involved.

922 As the information reported by CTAnalyser is rather limited, the categorical size map
923 image was used to calculate corresponding pore volume per size category. This functional
924 measure calculation on the size map image was achieved using own software.

925

926 **BoneJ (FIJI/ImageJ)**

927 BoneJ (Doubé *et al.*, 2010) is a freely available plugin module within the FIJI image analysis
928 platform which is a software distribution of ImageJ (version 1.47). BoneJ implements a
929 “structure thickness” measure based on MIB fitting along the medial axis, which is derived by
930 finding ridges on an Euclidean distance map (Dougherty and Kunzelmann, 2007). It results
931 in a map image of MIB diameters, from which a volume weighted distribution can be
932 obtained. The image histogram feature of FIJI can be used for this purpose, bearing in mind
933 that volume estimates are obtained by point-counting. As part of the investigative work of the
934 current study, it was determined that BoneJ version 1.3.12 (released 29th April 2014) and

935 earlier versions, produce MIB map images that do not conserve image structure.
936 Specifically, each MIB object generated within the map image overlaps the image
937 background, leading to inflated estimates of pore volume. As a result, each MIB diameter is
938 enlarged by approximately two image elements, which although small at diameter level, it is
939 sufficient to noticeably bias the estimated size distribution. This problem was corrected in
940 this study (and the subsequent BoneJ releases) by masking of the size measure image, i.e.,
941 setting to zero any measure that lies outside of the original object, as defined by the original
942 dichotomous image of pore versus solid.

943

944 **Quantim4**

945 Quantim4 (version 4.8) is an open source C/C++ function library
946 (<http://www.ufz.de/index.php?en=39198>) applicable mainly to Linux systems (the use of
947 features specific to the GNU g++ compiler mean that the code is not easily portable to other
948 systems). The analysis of images with Quantim4 requires some programming ability, but
949 owing to the convenient high level functions provided by the Quantim4 library a useful
950 analysis program can be both small and simple in structure.

951 Quantim4 uses a ball shaped structuring element in a sequence of morphological
952 operations guided by the Euclidean distance map. Morphological openings are applied to
953 individual image elements and these operations are both parameterised and ordered
954 according to a distance measure on those image elements (Vogel, 1997; Vogel and Roth,
955 2001). This approach is equivalent to the direct fitting of maximal balls, as described in
956 (Coeurjolly, 2012) and achieves results that are quite similar in practice. The underlying
957 algorithm can be briefly summarized as follows: first a distance map image is computed
958 using the squared Euclidean distance metric, then this distance map is used to construct the
959 “Open Map” (terminology provided by Quantim4 documentation) by mathematical
960 morphology. The final processing stage computes Minkowski functionals (Vogel *et al.*, 2010)
961 for thresholds of the Open Map, providing cumulative measures (including the volume
962 fraction of objects) per size interval. The size measure reported by Quantim4 is determined

963 as the diameter of a sphere containing a volume equivalent to that of the morphological
964 structuring element for each size class.

965

966 **DTM**

967 The method developed by Monga and co-workers (Monga *et al.*, 2007, 2009, Ngom *et al.*,
968 2011) involves a number of successive steps. The first consists of selecting pore boundary
969 points, defined as points in the interior of pores, which have at least one neighbor voxel that
970 does not belong to the pore space. A 3D Delaunay triangulation of boundary points is then
971 computed using the very fast code developed by George (2004). All tetrahedrons that are
972 not included entirely in the pore space are removed, and Delaunay spheres, i.e., spheres
973 passing through the four vertices of a given tetrahedron, are computed for the tetrahedra
974 that remain. These Delaunay spheres are maximal in the sense that they are fully contained
975 within the pore space and that no other sphere (within the pore space) contains it. The
976 centers of all the Delaunay spheres are then assumed to constitute the "skeleton" of the
977 pore space, referred to either as "medial axes" (Ngom *et al.*, 2011) or "Lambda-skeleton".
978 This approximation is reasonable because it can be shown that when the sampling of a
979 surface defining a volume shape tends to 0, then the set of the centers of Delaunay spheres
980 converges uniformly to the shape skeleton. The last step of the method then involves the
981 use of heuristic algorithms to compute a minimal set of maximal balls covering the Lambda
982 skeleton, with "minimal" interpreted in a cardinal sense. The basic idea of the heuristic is to
983 place iteratively the biggest ball, by maintaining a minimal covering with the already selected
984 balls. Once the minimal set of maximal spheres is obtained, the distribution of spheres can
985 be used easily to compute a pore size distribution.

986

987 **3DMA**

988 The 2005 release of 3DMA is an open source package consisting of many image analysis
989 algorithms invoked via a hierarchy of text menus (the 2011 or later release of 3DMA is
990 commercial software and provides a graphical interface; the commercial version was not

991 assessed within the present study). Analysis of pore size in 3DMA is based on detecting the
992 location of pore throats, these being the narrow apertures that separate a pore cluster into
993 distinct bodies. This is achieved by analysis of a “burn map” of the pore space obtained
994 using the pore space burning algorithm, which is equivalent to a form of distance map
995 obtained using either the Manhattan or chessboard distance metric (Lindquist *et al.*, 1996,
996 2000; Lindquist, 2002). The latter metric was used in this study. The discrete skeleton of the
997 burn map is determined using the LKC algorithm (Lee *et al.*, 1994) and then local minima of
998 the burn number (distance measure) on the skeleton are used to guide the search for
999 minimum area planar throats. The result of this analysis is a pair of binary encoded data
1000 files, one listing throat locations and the other body locations. In the former case the
1001 estimated area for each throat is also given, while the element (voxel) count for each body is
1002 given in the latter case. The distributions of these data can be plotted from within 3DMA, but
1003 we elected also to process the data files using our own software in order to control histogram
1004 binning.

1005

1006 ***DFS-FIJI***

1007 This method combines two software tools available within FIJI (ImageJ, version 1.47) that
1008 are both applied to the image of pore structure. The first tool, “Skeletonize3D” (Arganda-
1009 Carreras *et al.* 2008) generates a discrete skeleton map image of pore space using the LKC
1010 algorithm (Lee *et al.*, 1994). The second tool generates the Euclidean distance map of pore
1011 space, using an unspecified algorithm (nor is any author credited). The conjunction of these
1012 two map images (preserving the Euclidean measure only where the skeleton is defined) is
1013 referred to, within the present work, as a Discrete Skeleton Function (henceforth DSF), an
1014 approximate representation of the medial axis function (Blum, 1973). Discarding spatial
1015 information, the DSF can be interpreted as a population of local radius measures and hence
1016 may be used directly to form a population diameter distribution. Alternatively a means of
1017 approximating a volume measure per skeleton element is to treat each as being the centre
1018 of a disk, calculating the area of the disk and then extruding this by one voxel to obtain the

1019 volume of a circular cylinder. The total volume estimate obtained in this way does not
1020 resemble the true pore volume, but might be scaled so as to plot a crude estimate of volume
1021 fraction versus diameter. Irrespective of the manner of presentation of the DSF, imprecision
1022 is introduced by using only discrete information (map images) without reconstructing the
1023 underlying continuous medial axis function. Where the local pore diameter is even valued,
1024 the discrete skeleton map rounds the axial location to the nearest element, hence the
1025 selected Euclidean distance measure will be in error by ± 0.5 elements.