Dear editor, dear reviewers,

We thank the reviewers for the careful inspection of our manuscript and providing useful comments and recommendations that helped to improve our paper. We made effort to address all the reviewer's comments. Beside numerous corrections and further text, we added new Figures (Fig. 1E, F), a new table (Table 3), and another nine references. You find our answers in blue color. We highlighted the changes in the text by yellow background.

Anonymous Referee #1

General comments:

The analysis of pore space geometry in rocks is of great relevance to many areas of earth sciences. In this study, the authors propose to compare the results obtained from different pore space characterization techniques in two sandstones, the Bentheimer and the Rotthbacher.

Though I think this is a great topic to investigate, I find that the work itself does not bring significant value in its present form. A lot of research has already been conducted in this domain and it is difficult to see what new element the paper provides, aside perhaps from the spectral induced polarization part which I unfortunately had a very hard time following. Maybe the work could be augmented with a better review of previous findings and a more thorough extraction of information from the microCT images. In the abstract, the authors announce that they are going to characterize the pore space geometry, although nothing is done beyond acquiring and tracing various cumulative curves. In the abstract again, the concept of fractals is used but it is unclear (1) whether it is warranted to compute a fractal number from these curves as it is not demonstrated that they represent distributions of objects and (2) what the authors recommend what one should do with that value.

We thank for the critical remarks. We agree that a lot of research has been done in the domain of pore space characterization regarding µCT, MICP, and NMR. The main aim of our paper is to integrate an electrical method in this study. The spectral induced polarization (SIP) method provides a relaxation time distribution. It is assumed that the relaxation times are related to geometric length in the pore space. In order to verify this approach, a comparison with other methods providing insight into the distribution of pore sizes is necessary. The curves representing the cumulative volume as a function of pore radius provide the chance to compare the curves resulting from different methods. Additionally, the cumulative curves are used to assess the fractal behavior of the pores volume distribution. The fractal dimension is a useful number for up-and downscaling of geometrical quantities. Additionally, the fractal dimension is used in methods of permeability prediction.

Regarding the work that is done on images and the comparison between different data sets, I wish the authors had provided more information and figures on the image segmentation result as well as the result of the maximum inscribed sphere (MIS) computation. Also, the authors may be aware that such computation (MIS) can be used as a starting point for performing a digital equivalent to MIP which becomes then valuable to compare with the experimental mercury injection curve. In fact, a comparison between experimental MIP and digital MIP on one hand, and between MIS and NMR on the other would have made more sense.

Because the reader does not have access to the state of the images prior to the tracing of the cumulative ’pore size’ curve (MIS), it is very difficult to check whether the result is consistent. I have a doubt regarding the offsets observed in Figures 2 and 6 and I am wondering if what is plotted for the MIS is really the radius or rather a diameter. Please check. Also I am surprised to see that virtually no ’objects’ with dimension smaller than 20 microns was detected in either sandstone considering the image resolution of 1.75 microns per voxel and 1.5 microns per voxel for the Bentheimer and the Rothbacher, respectively.

As shown within figures 2 and 6, in fact pores in range of the image resolution have been detected and segmented. Nevertheless, these pores with small radii almost provide no significantly volume contribution concerning the results of the digital image analysis. We assume that this has a couple of reasons: first, the pore segmentation is greatly influenced by the density contrast between each individual phase (void and solid minerals), as well as by typical X-ray artifacts such as partial volume effects, which cannot be quantified in detail with conventional µ-CT. Additionally, the watershed algorithm will be sensitive towards the previously mentioned reasons and hence will lead to under-estimated volumes especially for small pores. We have addressed these effects within the revised manuscript.

Our study shows how other methods can be used to extend the resolution to smaller pores. We confirm that the pore radii are presented in Figures 2 and 6. The MIS data contains over 10000 points and it is a huge table so we only show the figures here. The data points of several voxels were eliminated because these data may either be pores or noise. We added a new Table 3 that compiles the minima, maxima and mean values of different pore sizes derived from µ-CT data of the two samples.

I don’t think that I can speak at length to the SIP part because I am not familiar with it. I would like to see a more intuitive explanation as to why it is appropriate to compare SIP data with a drainage (MIP) curve. Also I don’t understand how the data of Figure 5 on frequency-dependent complex conductivity is converted into relaxation times (assuming this is what is being done).

We recognize that SIP is a novel method in the field of pore size distribution. We added a paragraph to explain the Debye decomposition approach that is used to transform the spectra of complex conductivity into a relaxation time distribution.

In terms the organization of the paper, I found the figures confusing in the fact that they convey more information than is being discussed at first, forcing the authors to go back and forth when describing their results.

We have reconstructed the Results and the Discussion.

I am convinced the authors have at they disposal a great starting point to a valuable study. The sandstones picked are definitely materials of interest to the community and the high resolution microCT images can certainly be exploited further.

We recognize that the reviewer is most interested in the µCT images. It would have been possible to write an additional paper with special focus on the data processing and interpretation of the µCT data of the investigated samples. But the reviewer should consider that the µCT yields information, which is limited by the above described effects. The other methods are quite important to get insight into the pore space at the sub-micrometer scale. We investigated in our study how NMR, MIP, and SIP data can be used to get insight into the structure of pore size over a wide range of pore radii.

Other comments:

*I think the English could probably be improved (grammar and choice of words mostly)

We made some corrections highlighted in yellow color in the text.

*In the conclusion, an image resolution of 3.5 microns per voxel is quoted – please decide.

Correction done.

*I did not see the benefit of plotting the curves starting from the smallest injection radii - it puts an emphasis on the fraction where there is less data and also that likely contributes nearly nothing to flow, while dwarfing most of the important information.

We plotted the curves starting from the smallest resolved radii to show the different pore ranges detected from the different methods. The fractal nature of pore size can only be recognized if a wide range of pore radii is resolved.

*How were the injection steps chosen for the MIP curves? It seems nearly random, and also very sparse in the case of the Rothbacher.

The injection steps were automatically recorded by the equipment. In Figures 2 and 6 the MIP curves were plotted with step values of 10 (only every tenth value is shown) and 4 (only every forth value is shown), respectively, in order to show the data points clearly.

*Table 2 would be easier to look at if some mineral names were added to it.

We changed the term mineral phase to chemical components, and we add a reference describing the minerals of the Bentheimer sandstone.

*The resolution of the microCT images is great, the authors should be able to show much more detail at the grain scale. Have the authors attempted to determine whether the voxel dimension was a true image resolution?

Thank you for the positive evaluation of the µCT images of the two investigated sandstone samples. Certainly, the image quality is good down to a resolution of roughly two voxels. But this resolution is not sufficient to characterize the small pore space of sandstone underneath a distinct radii threshold as previously described.

D. Healy (Referee)

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General comments

This ms is a comparison of 4 methods for characterising pore space, especially pore size distributions. Data from tests on 2 sandstones are compared. Overall, it is a good idea, quite well presented, and should provide a useful addition to the literature. It’s easy to suggest extra tests or analyses but the 4-way comparison stands as is (but see below), and the separation into methods better suited for pore bodies and pore throats is good. I think this paper could be acceptable, subject to moderate revision. My main issues are with the relative lack of quantitative comparison of the methods, and the apparent underlying assumption of power law behaviour. E.g. in Figures 3 and 7, assuming you can make a case for power law behaviour, what is the departure of each line/method from a modelled power law prediction? The paper does not go anywhere near far enough in this regard, in my opinion.

We thank for the positive evaluation of the general idea of our manuscript with the focus on a comparison of different methods regarding their potential for a detailed consideration of pore size distributions over a wide range of radii.

Specific comments

Line 88-90 – much more clarity needed here; a log-log plot of these variables MAY show a straight line, which COULD then be interpreted as power law behaviour. Are you assuming a power law, and therefore fractal behaviour of the data? Perhaps a Maximum Likelihood Estimator approach might be relevant here (Clauset et al., 2009 SIAM Review; Rizzo et al., 2017 Journal of Structural Geology).

The reviewer is correct. The presentation of the cumulative pore volume as a function of pore radius as log-log-plot, which corresponds a power law behavior, is necessary to recognize the fractal behavior of the pore volume. A deviation from an idealized straight line indicates non-fractal behavior. We add the content of Maximum Likelihood Estimator considering the underlying assumption for power law behavior.

Line 106-107 – but this limit is rock/ CT scanner/segmentation dependent, right? So add the caveat, ‘for this study, the CT resolution limit is . . .’.

Correction done.

Line 165 – ‘arguable’

Correction done.

Line 199 –‘extent’

Correction done.

Line 231, 243 – ‘mainly’; can you be more specific about the modal proportions of minerals?
Correction done.

Line 240, 256 – what method for the porosity estimate?

Correction done.

Line 249 –'depositional'

Correction done.

Line 251 – 'shows'

Correction done.

Line 384 – 'extent'; 'differs'

Correction done.

Line 401 – 'of' not 'on'

Correction done.

Line 508 – Table 2; these are not mineral phases, they are chemical components!

Correction done.

Line 512/Fig 1 – SEM; what detector, BSE? Say so.

The detector is BSE detector.

Line 520/Fig 2 – I think we need to see 'raw' data for these methods; AND then the 'processed' data using the 'known' porosity. Let the reader judge the data.

Line 538/Fig 6 – as for Fig 2; let's see the raw data.

Figure 2 and Figure 6 are the 'raw' data, and Figure 3 and Figure 7 are the 'processed' data using the 'known' porosity.

Line 523/Fig 3 – my point above about assuming power law/fractal behaviour (NB, not the same thing) is borne out by these non-linear data. . .

Line 544/Fig 7 – these look quite non-linear; comment?

The whole curves in Figure 3 and 7 are non-linear, that is correct, so we only use an interval with a constant slope. We add the text in the Discussion: "The whole curves of the four methods are non-linear, and a Maximum Likelihood Estimator (MLE) approach might be relevant here (Rizzo et al., 2017). For example, in the case of the NMR curve of Bentheimer sandstone, the fitting of all data using the MLE reveals that the log-normal distribution is the most likely distribution with the estimated parameters μ=3.43 μm and σ=0.82 μm. These two scaling parameters are the logarithmic mean and logarithmic standard deviation, respectively".

Figure 7 seems an odd choice of final figure; perhaps add a sketch/cartoon of pore space, pore bodies + pore throats, and their distributions; mapped to the 'best' tools for quantifying them.

A. Bubeck (Referee)

Reviewers' Comments:

The submitted manuscript describes a study that aims to characterize pore size distributions, and the scaling of attributes, for two well-constrained types of sandstone using the following methods: 1) micro CT; 2) mercury intrusion porosimetry; 3) nuclear magnetic resonance; and 4) spectral induced polarization. Using curves of the cumulative volume fraction of pores and pore body radius, the methods are compared.

Generally speaking, the aims of the paper are clear, and data are well-presented. Improving our understanding of the nature of pore space in natural rocks is important to a broad range of study areas, including petrophysics and rock mechanics, for which a comparison of methodologies will be potentially very useful. It is my view, however, that the manuscript, in its present form, contributes little to our understanding of natural pore variability; resolution cut-offs for these techniques have been discussed previously. Many of the sections need rewriting to avoid overlap and repetition, and the motivation, and context for the study are unclear. For these reasons, it has been a challenge to review as thoroughly as I would like. Some simple restructuring and clarification could improve this but additional work is also necessary. For instance, the authors state early on that they provide a "multiple length scale characterization of pore geometry" - where is this data? A sentence in the conclusion section refers to "μ-CT enables a geometrical description of individual pore space. . .", but no data are presented. Providing additional geometric data would greatly improve the novelty of the work, and appeal to a broader readership. If the authors lack this data, I recommend that they remove references to geometry throughout. Furthermore, there is virtually no review of existing geological (or other) data derived using the techniques, nor their application, and a scattered discussion of the results towards the end of the manuscript. This needs work before it can be considered for publication.

We add a table (Table 3) with the geometric data derived from µ-CT in the Results.

The authors may decide that additional data, and/or background and discussion is beyond the scope of this work. If this is the case, then considering the broad readership of Solid Earth, they might consider submitting the work to a more subject-specific journal.

We selected “Solid Earth” because the pore space distribution and geometry is of interest for a broad readership of geoscientist: geologists, petrologists, geophysicists, petrophysicists. A comparison of different methods used in this field should be a motivation not only to apply the traditional standard methods. SIP is a novel method in this field. It is our aim to check the potential of this method in pore size characterization.

As a final note, the manuscript should be given thorough proof-read from a native English speaker; language, grammar, and sentence structure require work before I think it is ready to publish. I have not made specific edits related to these because they are numerous. Below, I have provided section-specific comments for the authors.

I hope the authors find them helpful.

We thank for the critical and helpful remarks. We tried to address the remarks and added additional information as recommended by the reviewer.

MS Specific Comments

0. Abstract The abstract reads fine and, with some minor edits, reflects the main findings of the manuscript.

Line(s) Comment 39 ". . .pore space geometry. . ." Global edit: you do not discuss pore geometry in this work. Please remove reference to it unless you have additional data to add.

We add the text defining the geometry here.

1. Introduction This section needs some restructuring, and expansion. For submission to Solid Earth, the work should mention the existing applications of these methods (geological, and others). The authors could consider including a background section to present existing uses of each method, their limitations, and how this data is used. This would improve the framing of the work, and broaden its appeal to different readers.

We add some references of the applications of these methods.

These are some questions I have from this section: a. What are your motives for the study? b. Who are you targeting with this work? Petrophysicists? Is Solid Earth the best place to present this? Porosity, and its effect on fluid flow and the mechanical behaviour of porous rocks is important to a range of study areas, and the subject of extensive study. A (brief) review of some the existing literature would help - e.g. recent uses of x-ray CT to analyse pore volume/geometry/distribution and fluid flow through porous media.

Line(s)

52-57 This isn't much of an introduction for the study.

We add the text defining the geometry mentioned in this paper.

63-67 I think you could move this to your discussion section and expand on how results from these studies compare to your own.

68-75 This section isn't very clear. You mention 3 separate published NMR studies but no others? It is not immediately clear what the relevance of these studies are to your own motivation, or results. You should add some background for the other methods too. Speaking for micro CT, there are several studies that analyse rock pores that the authors should take a look at, including: Lindquist et al., 2000; Ketcham, 2005; Nakashima and Kamiya, 2007; Takahashi el al., 2016; Schmitt et al., 2016; Saenger et al., 2016; Bubeck et al., 2017; Zhao et al., 2017 (this last one should be particularly helpful in helping you structure and present your work).

We reconstructed this section.

76-83 This section is partly repeated. Read through your introduction and keep it concise.

Correction done.

77 Either: add a section that deals with pore geometry specifically, and include additional figures/data, or remove this.

Correction done.

80 ". . .distributions are connected to each other. . ." This is unclear.

Correction done.

2. Methodology

This section is very long and it looks like a mixture of background and methodology, which could be split up. Keep your methodology simple: what did you do, and to what?

A number of questions need to be addressed in this section to improve the clarity of your results. a. How many samples did you use in total? b. How many samples did you run for each test? This needs explanation to demonstrate that the results are repeatable. You could add this data to table 1. You should also explain what size samples need to be. c. Does sample size effect the results for any of the methods? It should certainly be considered in micro CT. d. Did you characterize grain size? What role will this play?

We reconstruct this section and add the sample information in Samples and methods section.

Line(s)

91-99 This is background

Correction done.

99 The use of fractals (in geology alone) stretches considerably beyond these references. If you are presenting data on pore scaling relationships, expand this and move to a background section.

We change the term ‘geometric objects’ to ‘pores in sandstones and carbonates’.

100-102 Good. You should move your sample descriptions into this section.

Correction done.

103-108 Tell me what you’re doing/using before you tell me the limitations of it.

Correction done.

109-110 How have you performed this analysis, what were the results, and why is it best? More information and figures to explain this process are needed. Why compare with 2D SEM images?

We used a standard analysis of the µ-CT data. A comparison between 2D SEM and a 2D slice of the CT image is shown in Figure 1. We add a Figure showing 3D images of pore network resulting from µ-CT.

113 ". . . each individual pore. . ." Where are your data for individual pores? You could present data on the proportion of pore body radii (how variable are they within samples? How do your different sandstone types compare?), their geometry and preferred orientation (if present).

We add a new Table 3 summarizing the geometric information resulting from µ-CT. The data of individual pores contains over 10000 data points so the table is not shown in the paper.

114 Do you have references for this technique?

Correction done.

119 You're using MIP specifically for pore throat radius? Be clear about what data each method is providing!

131-132 Try to explain this more clearly.

The detailed explanation can be found in the reference. You can imagine the big pore as a room with a door (throat), and the MIP data records the size of the door instead of the room.

139-145 Background.

Correction done.

142 "a capillary model with cylindrical pores, of uniform radius. . ." - How useful is this? Is it reasonable to assume a linear relationship for natural pores, which may be neither cylindrical, nor uniform? Kleinberg, 1996 applies it to "slit-like pores". You should provide some geometric description of your pore space to support the use of this model. At the VERY least, explain how your results could vary. You could leave this as is here, but add a section to your discussion covering possible limitations of the method and expand on this there.

The reviewer addresses an interesting issue. The surface to volume ratio is not very sensitive to the shape of the pores. As mentioned in the text, a cylindrical pore results in a surface to volume ratio of 2/r and a spherical pore in 3/r. The resulting difference (factor 1.5) can be ignored when looking at a logarithmic scale. The word "uniform" is deleted. A cylindrical pore has a constant (uniform) radius per definition. The geometric descriptors are compiled in Table 3.

150-151 "The range of resolved pore radii depends on the used value of surface reflexivity." – You need to explain what value you have chosen, and why? If similar assumptions apply to the use of a single value here, address it in the discussion also.

The correct choice of surface relaxivity is not a simple issue. We have demonstrated in chapter 4 a procedure to determine *ρ* from a comparison of µCT and NMR. This procedure demonstrates a joint use of different methods improves the reliability of the derived parameters. Applying this procedure, we got different surface relaxivities for the Bentheimer (*ρ* = 54 µm/s) and Röttbacher sandstone (*ρ* = 237 µm/s).

175 Quantify the "restricted range"

Correction done.

182 Did you have any repeatability issues? Do you think two sufficient?

In most cases, two measurements are sufficient for checking the repeatability of the acquired complex conductivity spectra. If the two measured spectra show remarkable differences, additional measurements are performed until no temporal changes are performed.

187-224 This section is a mixture of background, discussion and results, with some repetition. Read through carefully and move elsewhere where appropriate.

Correction done.

3. Sample material This section is a mixture of methodology and results. Does it need to be a separate section? I think it would be helpful to have this information earlier. When you’re describing the methods, it will help to know what they are related to. The section could also be much shorter. I recommend you edit the information into your methodology, explaining the number of samples used and their sample sizes

Correction done.

Line(s)

227-228 Cut this. Unnecessary.

Correction done.

244-245 Remove 245-247 Background

We think that this additional information might be useful for the reader familiar with different types of sandstones.

251-262 This is your data? It needs to be in the results section.

Correction done.

4. Results The description of the graphs in this section is rather vague and unhelpful - you refer to "differences" between curves but don't describe or quantify these. Equally, stating that something is "larger" or "smaller" is also unhelpful. Quantify your observations!

I suggest changing the section titles here to refer to the parameters measured, i.e. pore volume fraction, pore throat radius, and scaling. Describe the results of these for each rock sample. This would be easier to follow than describing each rock; it is the technique that should be the focus of this section.

Correction done.

Your fractal data is currently lost in your discussion section. Move it into your results and consider a table that compares the dimensions obtained.

The reviewer finds all the derived parameters of our study in Table 1 including the fractal dimensions for the two samples.

Line(s)

282 ". . .wide range of pore radii. . ." State the range measured for each method, with

upper and lower limits clearly defined.

Correction done.

289 Explain this more clearly.

We describe in line 289 the observation in Figure 2 that the µ-CT curve is shifted to larger pore radii in comparison with MIP.

5. Discussion

This section is a confusing mixture of results and discussion. You should read through it carefully and remove results to earlier sections.

After reading the discussion, I have a number of questions that can be addressed by restructuring and expanding the discussion. a. What are the implications for the pore size distributions you have obtained? There is no real discussion of the importance of pore size distributions, or their use, in this work. . . b. What effect do you think your results have for studies of the mechanical behaviour and fluid flow properties of your samples? c. How do your results compare to existing characterisations of these samples - are you offering an improved resolution? How important is it for samples like this? d. How strongly dependent are your resolutions on rock type? How would they vary for other rock types: Limestones/volcanic rocks for example? e. Are the approaches described suitable for a range of rock types/sample sizes/porosities? I would like to see this section discuss the implications of the results more broadly. f. Which of the techniques is best? Which one provides the most, useful, information -and for whom? g. What are the limitations of your findings?

We reconstruct the Discussion section.

A summary table of the data provided by each method, and resolution would be helpful to readers.

We guess that the reviewer has overlooked Table 1, which is summarizing the parameters derived for the two samples of our study.

Also include a limitations section and use this as a basis to explain which methods should be used for certain applications, and how cautious researchers should we be when interpreting the results.

6. Conclusions A lot of the material in this section could be moved into the discussion.

Add a sentence summarising the importance of the result of this study.

Line(s) 399-407 These are (some of) your conclusions. The rest can be moved.

7. Figures/Tables

Suggested new figures: a. Demonstrate the 3D pore network would be helpful when describing your samples (e.g. Figure 3 in Zhao et al., 2017)

b. Segmentation/MIS

We added 3D images (Figure 1 E, F) showing the pore network for the two samples.

c.Plot your body radii against cumulative frequency.

We do not see the purpose of such a plot. We prefer to plot the cumulative volume as a function of pore radii as explained in the text, because this graph in a log-log-plot enables the determination of the fractal dimension.

Figure 1 No changes suggested.

Figure 2 Label the two values (0.238 and 0.184) in the caption.

Correction done.

Figure 3 No changes suggested.

Figure 4 Label each part as A and B; describe them separately in the caption.

The curves of the two samples are displayed with different symbols and can be easily differentiated.

Figure 5 No changes suggested.

Figure 6 Label values as for figure 2.

Correction done.

Figure 7 No changes suggested.

Table 1 Where does your permeability data come from? I recommend you convert these values to m2.

The gas permeability has been measured and Klinkenberg correction has been applied. The unit mD has been converted into m^2.

Table 2 No changes suggested.