Dear editor, dear reviewers,

We thank the editor and reviewers for the critical comments to our paper. We made effort to address all comments. You find our answers in blue color. We highlighted the changes in the text by yellow background.

Topical Editor Decision: Reconsider after major revisions (27 Aug 2018) by Michael Heap
Comments to the Author:
Dear Mr. Zhang,

Based on the concerns of the first set of reviews, I sent your manuscript for re-review. I now have two new review reports: one that recommends "rejection" and another that recommends "major revision". Their concerns centre around three issues: (1) missing references to key works, (2) insufficient discussion of the data and, importantly, (3) the absence of significant novelty. Because I think that these concerns can be remedied, I'd like to give you the opportunity to further improve your manuscript and resubmit to Solid Earth. I have therefore selected "major revisions". However, please be aware that I consider that addressing these comments will require large changes to the manuscript, rather than the addition of a few new sentences. If you are willing, please now prepare and upload a detailed point-by-point rebuttal letter and a revised manuscript.

Thanks,

Mike Heap (Topical Editor of Solid Earth)

We thank the Topical Editor of Solid Earth to provide us a further opportunity to improve our manuscript.
We have done much effort in addressing the comments and suggestions of the three reviewers in the first iteration of the reviewing process. Beside numerous corrections and further text, we added new Figures (Fig. 1E, F), a new table (Table 3), and another nine references.
Another recommendation of a “major revision” indicates that the improvements of the manuscript were neither fully acknowledged by the editor nor by the two reviewers of the second iteration of the review process.
Unfortunately, the remarks and recommendations of the two reviewers are not quite helpful for a further improvement of our manuscript. A general evaluation of the presented approach

is missing. The critical remarks of the two reviewers are related to minor issues of special methods and do not consider the integral inspection of data resulting from different methods with varying resolution, which is the main focus of our study.
Reviewer #1 recommends a rejection of the manuscript because he/she does not recognize the novelty of our approach and claims that the used Debye decomposition should be replaced by Warburg decomposition. We do not agree with the argumentation of this reviewer and address these issues in our "rebuttal letter" below.
Reviewer #2, who has already reviewed our manuscript in the first iteration, recommends further clarification of some issues that have already been explained in the text.
Nevertheless, we follow the recommendation of the Topical Editor and
(1) added eight more references to key works;
(2) improved the discussion of the data;
(3) indicated the difference of our approach in comparison to previous studies.
We hope that our additions will clarify the key messages of our study.

Report #1
The paper presents experimental data of different nature including NMR and SIP in order to characterize the pore size distribution of sandstones. In my opinion, there is nothing fundamentally new with respect to the work done by Niu Q. and Zhang C. 2017. Joint inversion of NMR and SIP data to estimate pore size distribution of geomaterials. Geophysical Journal International.

We are aware of this interesting paper of Niu & Zhang (2017). The reviewer should recognize that our manuscript does not propose a "joint inversion" of NMR and SIP data to get a more reliable pore size distribution. The paper of Niu & Zhang (2017) assumes that NMR and SIP resolve the same pore geometry. We and other authors (e.g. Revil et al., 2014) assume that SIP (and MIP) resolve the pore throat and µ-CT and NMR the pore body radius. Considering the different geometric parameter, a joint inversion can only be done if the ratio between pore body and pore throat radius it known. We spent effort in our study to get this ratio from the comparison of MIP and µ-CT data.

The second biggest problem I see in this paper is the use of the Debye deocmpostion which is fundamentally in error with respect to the basic physics of the problem. If my understanding is correct, it was shown that such a kernel inplies that if all the pore have the same size, the spectra are described by a Debye model. No metallic-free rocks is described by a Debyt. At best (very uniform materials), the transfer function is a Warburg (a Cole-Cole with an exponent c = ½ to be compared with a Debye for which c = 1). Using a Debye decomposition is therefore a huge mistake in recovering the pore size distribution. Why this has been described in many papers in the last few years since Florsch N., et al, Inversion of generalized relaxation time distributions with optimized damping parameter, Journal of Applied Geophysics, 109, 119–132, 2014., it seems that the authors are not aware of these works. I think they should spend more time in reviewing precisely the literature on the subject and redo the analysis with a Warburg decomposition instead of a Debye decomposition.

We thank the reviewer for addressing this issue, but we do not agree with this opinion. We know the interesting paper of Florsch et al. (2014) and acknowledge their work to generalize the decomposition of IP spectra using different kernels, but you will not find any statement in this paper that a Warburg decomposition is the only procedure to get a "true" pore or grain size distribution. This paper describes the mathematical and numerical procedures to get distributions based on different models.

There is a variety of papers that use the idea to overlay Debye models with different relaxation times to simulate IP spectra of media with a wider pore (or grain size) distributions (e.g. Leroy et al., 2008; Nordsiek & Weller, 2008; Revil & Florsch, 2010; Niu & Zhang, 2017). Please, have a closer look at the recent paper of Niu & Zhang (2017) which you mentioned in your review. Their approach is based on a Debye kernel as well (see their equations 5, 8, 12, and 13). I guess that no reviewer has recommended to reject this paper because of the used superposition of Debye models.

The paper of Revil et al. (2014), which has already been referenced in our manuscript, compares Debye (DD) and Warburg composition (WD) for a set of six sand samples. Regarding the peak and width of the resulting pore size distributions (but not the location), they show evidence that WD of IP spectra results in (a slightly) better agreement with the pore throat distribution from MIP. Having a closer look at their Figures 17 to 19, which indicate a rather good similarity between DD and WD results, this investigation needs "additional data … to confirm this finding" as stated by the authors. We recognize that for two of the six samples (436 and 499) a better agreement of the peaks of DD and MIP pore

size distributions. Therefore, we cannot recognize that the use of DD is a “huge mistake” that contradicts the physical behavior as stated by the reviewer.
Certainly, more studies have to be done to check different approaches. Our results, which are based on DD, demonstrate a fairly good agreement between the pore size distribution derived from SIP and MIP. A careful comparison between DD and WD (based on quantitative criteria and a large set of samples) is outside the main scope of this paper and cannot be done in three weeks given for the revision.

Other issues include a very poor review of the existing literature on these techniques in the introduction.

Considering the remarks of the reviewer, we extended the review of existing literature. We added more references in the Theory section, where the methods are described.

I also disagree with this statement “Rouquerol et al. (1994) reported that no experimental method provides the absolute value of parameters such as porosity, pore size, surface area”.

Unfortunately, the reviewer does not provide any justification for the disagreement. In order to avoid misunderstanding, we extent the text after this reference with remark to the fractal nature of the mentioned parameters.

I am also surprised that the earlier works by Slater and Lesmes are not cited.

One of the co-authors has got a successful collaboration with Lee Slater for many years and is aware of his earlier works with David Lesmes. The two authors published an interesting paper (Slater & Lesmes, 2002) relating the grain size (d_{10}) to the imaginary part of conductivity. The reviewer is correct that this paper may be regarded as an early contribution to combine geometric and IP parameters, but the idea to relate pore size distributions to relaxation time distributions was proposed later. We added this reference with others to acknowledge the early contribution.

No discussion is given for the small pore sizes that are hidden by the Maxwell Wagner relaxation.

The reviewer is correct that we did not mention explicitly the Maxwell Wagner polarization. We wrote in the text (lines 216 – 218):
"Considering that the complex conductivity spectra are affected by electromagnetic coupling effects or other polarization effects at higher frequencies and by a lower signal to noise ratio for lower frequencies, we focus on the frequency range between 0.01 Hz and 100 Hz."
We replace "other polarization" by "Maxwell Wagner polarization and dielectric effects". We add the following sentence:
"Smaller pore sizes are hidden by Maxwell Wagner relaxation and dielectric effects that are not easily related to pore geometry."

Report #2
Though I appreciate the efforts made by the authors to respond to my earlier comments, I still feel that this contribution is lacking essential material to support the results that are reported.

We have spent much effort to address the comments of all reviewers in our first revision. We have to apologize that we were not able to follow all recommendations of the reviewers. We paid attention to support the key messages of our study. We are aware that additional material can be included to clarify problems encountered by the different methods. But, we would like to present a concise manuscript that focuses on the key messages. A paper that combines the results of four different methods is not the place to provide and discuss details of each individual method. We preferred to provide a variety of references to the theory of the individual methods.

The assimilation of the MIP data to a pore throat size distribution on one hand and of the NMR data to a pore body size distribution is not substantiated.

A careful look into the text shows that we have provided an explanation for the different pore sizes resulting from MIP and NMR. We have described in the text of chapter 3 that mercury intrudes into the pores through the pore throats:
"Starting with low pressure, the pores with larger pore throats are filled with mercury. While increasing the pressure, the pores with smaller throats are filled. Reaching a certain pressure level P_c, a cumulative volume of mercury (V_{Hg}) has intruded into the sample that corresponds to the pore volume being accessible by pore throats radii larger or equal r_t according to Eq. (2)."

We have shifted this explanation into the Theory chapter and have added a remark to the new Figure 1, which shows a 2-D image of the pore space with pore throats and pore bodies.
We have added a remark in the Theory chapter related to NMR:
"It should be noted that the NMR method resolves the radius r_b that corresponds to the maximal distance to the pore wall. It can be represented by the pore radius of the largest sphere that can be placed inside this pore as shown in Figure 1."

The authors do not offer a single image of a pore space representing what they think the techniques are measuring.

Following the recommendation of the reviewer, we have added the new Figure 1, which shows a 2-D image resulting from µ-CT of the pore space with pore throats and pore bodies. Using this Figure, we explain which radius is measured by the different methods.

There is more than one way to generate distributions such as the ones showed for the pore body radius, and the very large contrast that is observed with the MIP results should warrant further investigation, starting with reviewing graphically how pore bodies are segmented and tagged. Then one would have to explain what these individual pores might have to do with the NMR signal.

We display the distribution of the pore body radius resulting from two different methods: NMR and µ-CT. The algorithm providing the pore body radius from NMR T_2 relaxation times is based on equation 5 as described in chapter 2. The uncertainty of the position of the NMR curve (in horizontal direction) is related to the unknown parameter of surface relaxivity. According to our experience, we determined the pore radius from µ-CT images using the largest sphere that can be placed inside each individual pore (maximum inscribed sphere method, e.g. Silin and Patzek, 2006, see new Figure 1). We agree that another approach would possibly result in a slightly different distribution.

As already mentioned in the previous review, I also think that the data set could be made more complete by adding an MIP simulation based on the images as well as a distribution of the maximum inscribed sphere radii in the pore space prior to pore separation.

We agree that it is possible to perform a MIP simulation based on the 3-D µ-CT data. Results can be found amongst others in Knackstedt et al. (1998). Considering the resolution of µ-CT (in this study: no pores considered with radii < 10 µm), most of the pore throats (< 10 µm) are not resolved. Therefore, an agreement between "synthetic" MIP data and measured MIP cannot be expected. A reliable MIP simulation requires sufficient resolution of both pore bodies and pore throats. In the case of our samples, we find that µ-CT resolves the largest pores. This is a strict limitation of the µ-CT method.

According to our approach, the cumulative volume distribution resulting from µ-CT can be "continued" to smaller pore radii after an adjustment of the NMR curve by selecting a suitable value for surface relaxivity. From a technical point of view, i.e. due to the DIA software used, pore separation is essential before performing the maximum inscribed sphere algorithm. Otherwise the "entire pore volume" will be approximated by one equivalent sphere. Please be aware, that the "in-situ" image is used for analysis, we do not perform pore network modeling in order to derive equivalent pore body diameters.

Reference:

Knackstedt, M., Shepard, A.P., and Pinczewski, W.V. (1998): Simulation of mercury porosimetry on correlated grids: Evidence for extended correlated heterogeneity at the pore scale in rocks. Phys. Rev. E, 58, R6923(R), 1998.