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Solid Earth Discussions

Interactive comment on "Calculating structural and geometrical parameters by laboratory experiments and X-Ray microtomography: a comparative study applied to a limestone sample" *by* L. Luquot et al.

Anonymous Referee #1

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General comments

The paper reports petrophysical, morphological and transport properties, such as porosity, pore size distribution and diffusion coefficient, of a limestone rock sample before and after a dissolution experiment. The different properties and parameters are measured experimentally in laboratory but also computed from X-ray microtomography images acquired on the same sample, before and after the dissolution experiment. The comparison between the two approaches (laboratory experiments and XMT image analysis) for the characterization of a limestone submitted to a reactive percolation

experiment is interesting. It is novel to my knowledge and should be published. However it is not always very clearly written and structured. Below is a serie of comments which should be addressed before publication, followed by a detailed list of technical corrections for the revised manuscript.

Specific comments

1. One major aspect of the study is the characterization (experimentally and using XMT images) before and after an experiment, as clearly written in 3296 / L26-29. I think this should appear in the title somewhere, maybe "a comparative study applied to a limestone sample submitted to dissolution" for instance. Indeed, the need to characterize the sample twice (after the experiment) prevents the use of destructive methods, such as mercury porosimetry for instance. Therefore the experimental protocole proposed in the paper to characterize the sample (which is then compared to the results from XMT images, in terms of values but also duration time) highly depends on this before/after characterization of a same sample.

2. Porosity determination with helium pycnometry is the most accurate and reliable method to quantify effective porosity. Helium is a very small molecule and can easily enter the connected pore network. Methods using water saturation have a higher degree of uncertainty (a full saturation with water is hardly met and depending on the pores type and size the sample can get superficially desaturated during the weighing procedure. In this paper, a water-saturated sample was required for the diffusion experiment and therefore the use of the "triple weighing method" (commonly called "liquid saturation and immersion method") to quantify porosity makes sense. However the possibility of using helium pycnometry should be mentioned in the discussion method, when comparing duration time for instance or when mentioning the source of potential error due to the triple weighing method. It is indeed fast and non destructive and the most accurate porosity measurement.

3. In the abstract and conclusion (3317/L2-4 and L17-20), the statements like "the val-

ues obtained by computing XMT images are in agreement with the classical laboratory measurements" or "provide similar results" should be qualified by adding "for the type of rock studied here". There will obviously be other type of rocks presenting a higher fraction of sub-resolved porous phase (or presenting a pore network efficient for fluid flow that cannot be resolved at all because the mean hydraulic diameter is far below the voxel size) for which it would not be the case and this is not at all discussed in the entire manuscript. The limit of XMT imaging mentioned in 3314/L9-15 should be much more developed and also discussed depending on the type of rocks investigated (voxel size versus pore size). For some tight rocks a laboratory experiment may always be much more accurate than any image analysis would ever be, with the existing technics so far.

4. The permeability should not be a part of this paper. The way it is announced in the abstract and in 3297/L5-6, the reader expects that it is measured and computed using XMT images and compared but it is not. Results are briefly given but they are not even discussed since there are no comparisons with XMT image analysis, which is the objective of the paper! I suggest to remove this part since it does not bring anything to the discussion. The aim of the paper, as it appears in the title and abstract, is not to discuss porosity and permeability evolution during the dissolution of a limestone rock sample. This has been well documented in the past already. For instance the paragraph 3311/L7-14 could be reduced to one sentence stating that different types of dissolution pattern were observed in previous dissolution experiments and that XMT images can bring insight into the dissolution process and allow visualization of a wormhole in the present case. Dissolution patterns could actually also be anticipated from the Peclet and Damkohler number (Golfier et al., 2002), but then again, this is not eat all in the scope of the present paper.

5. Other parameters are mentioned and listed in Tables but not discussed at all or not even mentioned in the results parts, or the opposite. I suggest they should either be removed everywhere, either be fully treated as the other parameters (that is describe

the method, describe the Figure, clearly state the results and discuss the results). This concerns BET, S/V, tortuosity and skeleton.

- Nothing is said about BET measurement in materials and methods, and nothing is said at all about either BET nor S/V from XMT images in the results and discussion. They are listed in Table 1 but never mentioned nor compared. - Tortuosity is mentioned in the abstract and in the method section but then never mentioned anymore in the rest of the paper. - There is a subsection explaining how to extract a skeleton in materials and methods although it is not really mentioned what it will be used for or compared with, regarding laboratory experiments. And then at the very end of the discussion 3316/L10-13, the authors said that it gives the best pore and throat size distribution and refer to a Figure that has never been mentioned before in the result part, nor discussed at any point before this last sentence concluding on pore size distribution, a topic which represent a high fraction of the manuscript interest.

6. The title of the section 2.1.1 "rock sample analysis" might not be the most appropriate since most of the paper is about rock sample analysis. I suggest to just use "rock sample". This subsection mentions porosity values. Where do they come from? If it is a result, it should be in the result part and if it is not then details or references should be given. I am assuming that it is the values mentioned 3307/L5 "higher than the one provided by the quarries miner companies". Is there additional information on these measurements, such as the method used or the sample volume?

7. It would make the materials and methods section more clear and the general manuscript easier to follow if there could be a Table listing all the properties and parameters and how they are estimated (measured in the lab? (if so which method?) also computed from XMT images? Or just one or the other?). Also, concerning the XMT images section (2.2), the chord length distribution functions are introduced under geometric parameters, which makes sense at first. However, in the XMT results section the results concerning chord length are listed first for the pore size distribution analysis. This may be a bit confusing. I would suggest starting the pore size distribution

section (in materials and methods 2.2) with a sentence explaining that it was analyzed on the identified resolved pore space using two different methods: maximum inscribed-spheres and chord length and then detail both in that same section. And what about the skeleton then? It is also written explicitly 3304/L18 “using a Voxaya module”. Isn’t it the case for most analysis, as mentioned at the beginning of the subsection 3302/L18-19: “The processed images and results were mostly computed with Voxaya’s software”? Finally, subsection 2.2.2 could benefit from a sentence listing on which phase was applied the different algorithms (and this information could also appear in the suggested Table). It is not always clear. For instance I assume that the diffusion experiment was simulated on the connected resolved porous phase (it is never mentioned). What about the pore size analysis? Was it performed on the complete void phase or just the connected fraction of it? It needs to be clearly stated at one point.

8. Following up on the organization and structure of the manuscript, I would also recommend using the same subsection titles for the results section concerning the laboratory experiments and the XMT analysis. I suggest for instance: porosity evolution, pore-size distribution, diffusion coefficient (note the use of singular in the titles).

9. Equation (1) should be presented as the Young-Laplace equation and the assumptions behind this equation (cylindrical approximation) could be reminded as it may help the discussion when comparing the different approaches for the pore-size distribution. It would also have been interesting to compare with pore (throats) size distribution extracted from a Hg porosimetry test performed on a sample cored aside for instance. If the data are not available, this could at least just be mentioned in the discussion part in the paragraph 3316/L1-3. And there is a recent paper by Yang et al., 2015 that could be referenced there: Yang, F., Hingerl, F., Xiao, X., Wu, Z., Benson, S. M., and Toney, M. F., 2015. Extracttion of pore-morphology and capillary pressure curves of porous media from synchrotron-based tomography data, Scientific Reports, doi:10.1038/srep10635.

10. In the subsection “connected porosity” (3304), it could be mentioned that the resulting pore space is the effective porosity which can be compared with the one measured

experimentally.

11. The calcite dissolution equation (R1) is not a result and could be introduced in subsection 2.1.3 for instance.

12. Subsection 3.1.1 reports that 4 measurements were performed. I am assuming it is four times on the same sample (before, and then after dissolution). This is not clear. Also it should appear in the materials and methods section as it refers to the procedure more than the results itself.

13. Avoid “slightly” or any subjective terms. If you want to highlight the “slightly higher increase in porosity’ (3308/L14), quantify it by writing “a higher increase in porosity by . . .%”, or just write “an increase in porosity”. 3309/L4: “slightly” again and 3309/L5: “a small decrease” (how small is small?). 3314/L20-21: “the estimated porosity is quite similar even if the difference is more important”, please quantify and/or rephrase.

14. 3309/L5-6: This sentence is not clear and this is also the first time that pore clogging is mentioned. Rephrase and provide maybe a bit more detail on this statement.

15. In subsection 3.2.1, the estimation of uncertainty on porosity calculation is done for the resolved phase. It would have been interesting to do the same for the sub-resolved phase. Also, in Table 2 or in the text, it would be great to list the resulting porosity value when moving the threshold value to + or - 1 or 2.

16. In section 4, the two approaches (lab and XMT) are compared in terms of duration. What about the cost and accessibility (having rare beamtime at a synchrotron facility versus having permanent access to a laboratory equipment)? Maybe few words could be said about these aspects too.

17. The paragraph discussing diffusion coefficient should be better structured. The comparison of the values reported in the results section should be written first before mentioning that for both cases the diffusion coefficient increases after dissolution. Also the last sentence should be balanced by adding that it is the case for the studied

limestone (or a similar rock presenting similar pore structure and morphology). Indeed, a sample presenting a high fraction of sub-resolved porous phase might bring much more discrepancy between the laboratory experiment and the XMT analysis. It could actually be mentioned that there are less differences between Deff(I-) and Deff(XMT) after dissolution when the pore network has a well developed wormhole and a generally better connected resolved porous phase.

Technical corrections

3294 L23 / "...rock pore structure..." L25-26 / "Porosity is a key petrophysical parameter indicating..."

3295 L1-2 / "...differentiate the total porosity from the open connected or effective porosity, which is the fraction of porosity accessible by any fluid." L3-4 / "...and tortuosity allow to quantify the ability to extract or inject fluids." L21 / "...important to better understand these issues." L25 / replace "realized" by "performed"

3296 L26 / "to explore" L27-28 / "between variables computed from ... and those traditionally measured..."

3297 L9-10 / "Quantifying the pore network characteristics of a same sample...methodology to two different pore networks and enhance..." L19 / "...(CaCO3) and is named Bvl in the paper. This limestone is commonly referred to as Beauval rock and is coming..."

3299 L3 / "...acquire twice the retention curves..."

3300 L22 / "...solution by mixing..."

3302 L3 / "X-ray microtomography images were acquired on the ..." L9 / 3-D reconstruction: which software? (provide brief details and reference) L11 / replace "around" by "about" L15 / "...quantify the volume and morphology of the pore structure identified during the segmentation process. Using the 3-D..." L17 / "...and connected porosity and geometrical properties..." L19 / "...was applied to both images."

3303 L1 / reverse the sentence: "segmentation is one of the most important step in image analysis." L2 / "...voxels that belongs to...assigning them a..." L3 / "...solid phase which have the highest..." L4 / "...phase referred to as void phase..." / "We name subresolved..." L10-11 / I suggest you use quotation marks for microporous phase and macroporous phase. L14 / delete "the": "...computing porosity..." L16 / "A first segmentation..." L18 / replace "realized" by "conducted" or "performed"

3304 L12 / "...identifying neighboring voxels L14-15 / "...both resolved and subresolved porosity." L17 / "...quantify pore network characteristics such as pore size." L22 / "...each voxel of the pore space to the interface..."

3305 L18 / "...a Brownian motion, the diffusion..." L19-20 / Is it a sentence? Rephrase if possible.

3306 L22 / replace "primordial" by "essential" L23-24 / choose between "for example" and "among others"

3307 L1 / delete "classical" L2 / "...and give us..." L7-8-9 / "After the dissolution experiment" (there was only one experiment, right?) "the same methodology was applied four times and we measured a final porosity...". L10-12 / "Mass balance calculation from the dissolution experiments were performed using Eqs. (5) and (6) and the final porosity was evaluated using the initial porosity..." L15 / "...connection of initially non-connected..." L16 / "...connected porosity resulting from the dissolution..." L22 / "We measured the retention curve by draining the sample both in the flow and counterflow direction in order to evaluate the pore size anisotropy." L26 / "...gradient of capillary pressure..."

3308 L9-10 / "Moreover some heterogeneity can be observed along the sample..." L13 / "The results show that after dissolution, the amount of pores..." L16-17 / "...the second major difference between the retention curves before and after dissolution appears..." L19 / "...radii are present through..." L21 / "The results also display some differences in the pore-size distribution..." L24-25 / "They are however minor when

compared with other experiments. . .” L25 / “Most discrepancy is visible. . .”

3309 L1 / replace “smaller” by “lower” L2 / “. . .that dissolution also occurred in these pores. . .” L3 / “However, the water contents. . .” L13 / “. . .diffusion coefficient, Deff(I-), listed in Table 1. After the dissolution experiment, the diffusion coefficient is increased by one order of magnitude, as suggested the noticeable incrase in the slope of iodide increment after dissolution.” L19 / “coefficients and tortuosity, as well as their evolution with dissolution, are coherent. . .” L23 / “The change in the sample permeability with time k(t) induced by the dissolution experiment is reported (. . .) due to calcite dissolution, as previously. . .”

3310 L1 / “. . .8h, which is surely associated with the breakthrough. . .” L5-6 / “The total porosity calculated from the XMT images, on sample Bvlbe (before dissolution) and Bvlaf (after dissolution) is 17.13% and 22.80%, respectively.” L13-14 / “. . .indicate that the smallest threshold values were used to estimate. . .” L19-20 / “. . .images obtained by setting the threshold value to n.” L24 / “. . .characterized by a high amount of large pores”. Note: by the way, this is not shown in Table 1. Is this a mistake?

3311 L2 / “. . .both resolved and subresolved porosity. . .” L4 / “. . .increases to a higher extent. . .” L6 / “. . . is higher for the first. . .” L18 / “Table 3 indicates the volumes. . .” L26 / “. . .compose” (space) “the percolating. . .” L26 / replace “higher” by “larger”.

3312 L1 / “The pore size distribution of the resolved porous phase was calculated for sample. . .”. Note: pore-size and pore size, both are used in the manuscript, choose one. L6 / “. . .also estimated. . .by performing. . .” L12 / “Figs. 3 and 8 show that. . .” L16 / “. . .to intermediate pore diameters. . .” L18 / “. . .process. A significant amount of pores presenting diameters comprised between 1 and 3 mm are measured and pores having a diameter up to 20 mm in the x direction can be found. It corresponds to the local. . .” L24 / “With the Psd analysis, the highest pores. . .”. “highest”: do you mean “largest”, or “most commonly found”? Eventually rephrase that sentence to make it more clear-cut.

3313 L13 / “This section compares the different. . .” L14-15 / please rephrase “before

considering these parameters”, this is too vague. You could also simply start with “We first compared. . .analysis duration for each parameter. . .” L17 / “The complete image processing. . .” L21 / “. . .using image processing. . .”

3314 L7 / “. . .part of the initially closed porosity. . .” L9 / replace “nonetheless” by “however” L9-10 / “. . .the high dependence of all parameter on the voxel size. . .” L17 / “. . .between the data computed from XMT images and the ones measured experimentally.” L18-19 / “. . .method is only different from the effective porosity extracted from XMT images by 0.74%.”

3315 L2 / replace “is” by “represents” L4 / “Regarding porosity measurement and analysis, we can conclude that porosity calculated. . .is the most reasonable. . .” L8 / “The same conclusion cannot be drawn for the pore. . .” L10 / “Fig. 3 displays the pore size distributions. . .” L17 and 18 / replace “larger” by “higher”.

3316 L4 / “To summarize, one should note that the pore size distribution. . .curves” (no coma) “indicates. . .” L14 / “measured experimentally and using XMT images is the effective. . .”

3317 L1 / conductivity ? L23 / “Futher developments using the extracted skeleton will also allow to. . .”

Table 2 / “Sensitivity analysis of the threshold value n on the resolved phase volume. . .”

Figure 9 / “Porosity change along samples. . .” This Figure concerns diffusion, shouldn’t it be somewhere in the title?

Interactive comment on Solid Earth Discuss., 7, 3293, 2015.