

General comments:

This is an interesting paper and addresses the importance of iron oxides on NMR signals, in this case focusing on T1 relaxation. And authors also probed into the relationship between surface relaxivity  $\rho_1$  and iron content. The structure and organization of this manuscript is good, and the presentation of the data is also satisfactory. The authors covered a lot of topical areas: impact of paramagnetic materials, novel NMR relaxation analysis, and so on. I feel a bit lost about the focus and the main findings of this paper. There are couple of other issues and suggestions

1. Pore size distribution estimation from particle size distribution is not reliable. The NMR mode analysis is based on the assumption of narrow (single) pore, I feel it is difficult to be convinced for this particular experiments as iron oxide precipitation would generate much smaller pores. This is a crucial point as the authors use reff information intensively, including calculating the diffusion regime. The updated reff could significantly alter the results and interpretation. Additionally, surface area analysis (i.e., BET) could help authors answer few ambiguous observations, e.g., the difference in surface relaxivity between goethite and ferrihydrite.
2. Can the authors discuss on the choice of coarse grain size particles? Also discuss what if the particles are fine.
3. In the study, only T1 relaxation has been studied (T2 was only used to calculate porosity). T2 relaxation is more important and it would be necessary to conduct T2 experiments and analysis. If both T1 and T2 measurements are obtained, more parameters like  $\rho_1/\rho_2$  can be extracted to provide insights of NMR monitoring of iron oxides. Why the authors didn't consider using low-field NMR core analyzer instead of one-side NMR-Mouse?
4. Similar to the first comment, the hydraulic conductivity should be measured in the lab to compare with NMR estimated value (from equation 12).

Specific comments:

1. Intro – The significance of studying iron oxide in saturated porous media is beyond the control of negative incrustations. I suggest authors consider making a broader argument of the importance of such study.
2. Intro – line 16 to 17 on page 2. The introduction of applying geophysical methods seems too sudden. The aim of this study would be better to placed after the introduction of NMR relaxometry. I think the effect of iron oxide (or paramagnetic materials in general) on NMR (surface relaxivity) needs to further reviewed, and more references should be added here.
3. Basics of NMR – line 16-17 on page 5, I didn't follow how to simplify  $\xi_n$  to  $(n + 1/2)^2\pi^2$ . Can authors further explain (use formula if applicable)/
4. Basics of NMR 2.4 – Do the authors assume single dominate pore size in analyzing the data? Can authors elucidate the applicability of Müller-Petke et al., (2015)'s conclusion in this study? For example, what characteristics of the samples used in this study to make this single pore size assumption valid?
5. Basics of NMR 2.4 – Did you do similar intensity and  $\rho r/D$  simulation and parameter search for T2 relaxation? Does the same conclusion hold?
6. Page 6, repeated use of the word 'unambiguous', consider changing some of it to other words like 'nonunique'.

7. Basics of NMR 2.4 – Could the authors define what are apparent surface relaxivity and apparent pore radius? Equivalent value or NMR estimated value? The last sentence of this section ‘An important objective of this study is the comparison ...’ seems to be a bit lost in the context. If this is an important objective, I suggest the authors review the relationship between  $r_{app}$  NMR and  $r_{eff}$ .
8. Material and methods – I suggest the authors use a flowchart to facilitate the explanation of the sample preparation and iron coating treatment. Why the authors didn’t measure the  $r_{eff}$  using MICP or imaging analysis? The estimation of  $r_{eff}$  from particle size is not reliable. If the authors want to compare the  $r_{eff}$  with  $r_{app}$  NMR, a realistic estimation of  $r_{eff}$  from analytical characterization is necessary.
9. Material and methods – line 18 page 8. ‘due to the high proportion of quartz, contents of siliceous iron are generally expected to be very low in fresh filter sand’. Does it mean the siliceous iron content is extremely low due to high purity of  $SiO_2$ ?
10. Material and methods – 3.4 why  $B_0$  has a strong gradient in  $z$  direction? Inhomogeneities in permanent magnets? Could you elaborate on this? I’m curious to know.
11. Results and discussion – 4.1 page 11 line 22 and line 30 ‘the latter exhibits a relaxation time of less than 0.2 s’, it didn’t seem to be 0.2s to me from the figure. Why coarse material will contribute to uncertainties in porosity estimation?
12. Results and discussion – 4.2 What is the scanning interval in your experiments? I thought you use 8 measurements at different depths for each sample, but the data points on figure 5 look much more than 8.
13. Results and discussion – 4.2 ‘This assumption is acceptable because the grain size distribution and consequently also the pore size distribution is narrow for the well-sorted materials studied here’ This statement is not convincing. I would expect a quite broad range (at least bimodal) of pore size distribution as much smaller iron oxide precipitation occurred. Especially authors also pointed out that  $r_{app}$  gets smaller when iron content increased. As I brought up before, the estimation of pore size distribution from grain size distribution is not convincing and the authors need to show evidence of pore size distribution from analytical measurements.
14. Results and discussion – 4.2 Did the authors calculate  $K$  using other models like SDR or Coates model? How did it compare to the  $K$  estimation using equation 12? Which equations you used to calculate  $K_{KC}$  and  $2.20 K_{Hz}$ ? Did you actually measure  $K$  in the lab for different samples? It is very necessary to do such measurements.