

General Comments

The study aims to demonstrate an exit effect (outer filter) in solute transport in a soil column with artificial macropores. They utilize advanced methodologies such as breakthrough experiments, MRI, and Comsol Multiphysics software. The results showed that an exit effect occurs with a good agreement between the three independent methodologies. I agree with your findings and congratulate the researchers for the hard work. Indeed, the study shows an exit effect on solute transport. Therefore, the overall research question is answered.

Before publication, I suggest an in-depth English edition. I found some technical corrections (see below); however, I am sure that I drop some more. Finally, I would like the authors to answer the next general and specific comments.

Q1. You did a nice introduction to the problem. You highlight that column laboratory experiments are commonly used to study the transport of various contaminants in soils or fit experimental data with a transport model. Considering the second highlight (fit experimental data), we commonly fit the dispersion length for a non-reactive solute with BTCs without considering exit effects and then use it for model simulations. Given your results for columns B and C. Do you think that the magnitude of the fitted dispersion length using the blue or red curves in Figure 3, might be strongly different?. I mean, we should be worried about the current estimation of dispersion length without considering exit effect under laboratory conditions?. I think the discussion about this is important in your manuscript because you also mentioned this in the conclusions. You indeed found an exit effect with your study, but it can be irrelevant for the fitted parameter (e.g., dispersion length). Finally, it could be interesting to see the exit effect for an adsorbing solute with COMSOL to expand this study (optional).

Q2. You demonstrate an exit effect using a soil system with a high macropore diameter (3 mm) in a sandy matrix with a mean pore diameter of 0.58 mm (for some researches, that diameter is indeed a macropore itself). The 3 mm macropore connects perfectly to the top and bottom boundary of the column with the filter. Therefore, your system allows fast water flow and solute transport in both domains, which can be different from the standard description of macropore flow where the soil matrix has very low permeability, and macropores can terminate at different depths. Thus, I think your system is convenient for observing the exit effect because of the high differences in permeability between the filter and the bulk soil system. What happens with the exit effect if the macropore terminates in the soil matrix before reach the bottom filter? What happens with smaller macropore diameters where the flow velocity is lower than for 3 mm macropores?. These questions are important for analyzing your conclusions. Please check your conclusion from line 371 onwards; they cannot be general because your study is particular, perhaps, unrealistic.

Q3 Finally, in your conclusion (Ln 371), you mentioned that “Our results shows that this knowledge is crucial for the understanding of the outcome of transport experiments in heterogeneous columns and for the accurate inference of transport properties from breakthrough studies.” As I mentioned in Q1, I think you did not demonstrate the last point. We do not know if the exit effect is critical for the accurate inference of transport properties from breakthrough studies with your study. From your study, we can know that the exit effect

happens in your particular setup (which is probably deviated for common heterogeneous soils with macropores).

Specific comments

Abstract: Please mention that your study is only under saturated conditions.

Ln 36 What do you mean by “real soils.”?

Ln 39. Please explain what you mean by this sentence, “the effect of macropores being expected to culminate when they are activated (i.e., water-saturated)” The sentence is tough to understand.

Ln 63 Please include the error in the construction of the cylindrical macropores generated by the 3D printer. Commonly for smaller diameters, the error might increase considerably.

Ln. 45. In the sentence related to the objective, please state that the soil system was under saturated conditions. The breakthrough experiment can be done either in undersaturated or saturated conditions.

Ln 110. You mentioned that MRI is utilized to visualize water flow and Gd^{3+} transport. However, in this section, you only mentioned that MRI was used to visualize the latter. The transport of Gd^{3+} can be somewhat different from the water flow due to dispersion or diffusion. Did you visualize water flow or just Gd^{3+} ?. In the results and discussion, you barely mentioned water flow also. Please check.

Ln 141 You mention “Stokes equation”. However, I would like that you mention the specific equation that you are using. I might think that you are simulating Creeping flow or Stokes flow in your macropore for fully saturated conditions. This kind of solution is special for low Reynolds numbers. Finally, I think you drop the gravity term in Eq. 2 (please check) because you simulate in the vertical flow direction.

Ln 151 Why are you not considering dispersion in the macropore?

Ln 167 How did you compute the new mean pore diameter?

Ln 170 Please specify if that porosity was obtained from the factory or you guessed it?

Ln 172 This equation was computed under laminar flow conditions? If so, please mention it.

Ln 174 why are you using a value different from 0.57 mm for the dispersion length?

Ln 176 Do you have a wall boundary condition for the 0.5 diameter openings that connect the macropore with the matrix?.

Technical corrections

Title: I think “macropored” is not correct. Perhaps macroporous.

Ln 2. “Macropored porous media” do not sound okay to me. It could be “filled with macropores.”

Ln 2-3. I suggest to change “numerically investigated, **and the**” by “numerically investigated. **The**”.

Ln 4. I think you can delete “of the presence of.”

Ln 5. I suggest you modify “macropored systems” by macroporous systems. Please check yourself all the times that you mentioned “macropored.”

Ln. 16. I suggest deleting “Broadly speaking.”

Ln 24. I suggest removing “presence of the”

Ln 30. Check that you used “and” several times. Perhaps you should remove the “and” in “and frits.” Alternatively, rewrite the sentence.

Ln 33. Please considered change “may trigger disturbance of water flow” into “may trigger water flow disturbance.”

Ln 36. Please considered to split this sentence into two “Real soils frequently contain macropores (Beven and Germann, 2013), which are large and continuous openings known to be involved in the rapid displacement of water and chemical substances, and various breakthrough experiments have been performed to study the role played by single macropores embedded in porous medium (Allaire et al., 2009)”

Ln 39. Please check the next sentence because is hard to understand “Unsaturated conditions being difficult to sustain in a well-controlled fashion, and the effect of macropores being expected to culminate when they are activated (i.e., water-saturated), many results have been obtained from macropored columns operated in the saturated regime, with different artificial systems: packed soils containing constructed macropores, macropored sandy media, glass bead packings crossed by a macropore, etc. (Allaire et al., 2009; Li and Ghodrati, 1997; Ghodrati et al., 1999; Lamy et al., 2009; Batany et al., 2019).” I think you should not use “being” e.g. “Unsaturated conditions **are** challenging to sustain in a well-controlled fashion. Also considered to split and rephrase the sentence is too long.

Ln 45. I suggest removing “of the presence.”

Ln 58. I suggest modifying “Finally, the sand was dried at 105 °C during 24 h and” into “Finally, the sand was dried at 105 °C for 24 h and.”

Ln 76 In the next sentence “for some of the experiments”, please remove “of the”.

Ln 114 In the next sentence, “Due to its paramagnetic properties, Gd_{3+} is known to be an excellent MRI contrasting” please remove “known to be”.

Ln 125. Please consider removing “as” from the next sentence “as the measured 16 echoes.”

Ln 126-129 Please use the next sentence “ Moreover, due to the short recycling delay used to keep measurement time below 4 min, the resulting signal depends simultaneously on the spin-lattice relaxation time T1 and the spin-spin relaxation time T2, thus complicating quantification of a simple comparison with a reference. The MRI images can nevertheless be used to evaluate where Gd_{3+} is present within the column.”

Ln 136. Please remove “one” from “another ONE for the macropore.”

Ln 145 Please remove “in” from the next sentence “the surrounding porous medium and in the filters”

Ln 151 I suggest to modify “The transport of the non-reactive solute was modeled” into “ The non-reactive solute transport was modeled”.

Ln 155 Please remove IN as before.

Ln 169 Please use the next sentence (included “a”) “The filter was modeled as a thin porous slab.”

Ln 174 Please correct “longitudinal.”

Ln 179. In the next sentence “of injected solution and to 0 afterward” it should be “afterward” and please remove “to”

Ln 205 I suggest modifying “The decrease of” into “The decrease in”.

Ln 209. In the next sentence, “hollow cylinder used in column B with a plain one to investigate” replace “by” for “with”.
”

Ln 211 In the next sentence, “the presence (blue curve) and in the absence.” please remove “in”

Ln 214 In the next sentence, “Thus, the solute having entered the column” please replace having entered by entering.

Figure 3: In the next sentence, “the presence (blue curve) and in the absence” please remove “in”

Ln 217 Please consider changing your sentence for this one “This is not the case for column B, whose macropore is perforated; thus, it can experience some solute transfer between the macropore and the surrounding matrix”

Ln 230 Please modify “These images were taken over the course of the injection” into “These images were taken throughout the injection of”

Ln 271 Please considered modifying “which is its influence on the position of the second peak” into “which is its influence on the second peak position”

Ln 273 Please consider to change “On the downside, the modeling of the first peak” into “On the downside, the first peak modeling appears challenging”

Ln 274 Please change “we did not succeed...” by “we failed to reproduce this portion of the experimental BTCs quantitatively.”

Ln 244 Plase modify the next sentence, “We start the discussion by the case” into “We start the discussion with the case”

Ln 287 I think you should change “Afterwards” by “Afterward”

Ln 293 Please change “on the way” for “how” and “transfered” by “transferred”

Ln 301 Please modify “nearly horizontal until its gets sufficiently” into “nearly horizontal until it gets sufficiently”

Ln 372 Please consider modifying “Indeed, when it comes to fitting the experimental data, a good knowledge” by “Indeed, when it comes to fitting the experimental data, good knowledge”

Ln 331 Please modify “field were obtained at the inlet and at the outlet of the column” by “field were obtained at the inlet and the outlet of the column”

Ln 339 I think you should remove “one”

Ln 343 This sentence is too long, considered split perhaps in “whereas it” by “.In contrast, it”... “However, despite the symmetry of the streamlines between the inlet and the outlet of the column when filters are present at both extremities, qualitatively, solute transport seems to be rather unaffected by the presence of an inlet filter, whereas it is strongly impacted by the presence of the outlet filter: images acquired by MRI (Figs. 4b and 4c) and computer simulations (Figs. 5b and 5c) show that the solute front is nearly horizontal in the lower half of a macropored column containing a perforated macropore”

Ln 347 . Please modify “The presence of the inlet filter probably alter” by “The presence of the inlet filter probably alters”

Ln 380 Please modify “This issue requires a careful consideration of the potential impact” by “This issue requires careful consideration of the potential impact”