Technical note: The use of an interrupted-flow centrifugation method to characterise preferential flow in low permeability media

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Third response to reviewer's comments

Comment from reviewer #1

I think the paper should be acceptable for publication following further revision. The answer to review #1 clarifies that the authors are presenting only an 'empirical methodology' and that the analysis is there only for secondary 'complementary' support. The addition on lines 387 to 399 is quite useful as it shows that a dual model is not necessary for an experiment without interruption. They must add another numerical experiment showing that for an interrupted experiment and not using the dual model they cannot fit their data incidently hysteresis would lead to results similar to their dual model I believe].

In their reply they also say'the whole breakthrough curve was fitted, not just the rising limb but also the decrease during the resting phases'. However it seems they only presented the experimental data for the rising limbs, they should include the data for the resting phases in their revision as well.

Response:

Many thanks to the reviewer for the suggestion for this additional numerical experiment which has been very useful in strengthening our argument. We have added the following text after the discussion of the first numerical experiment the reviewer suggested in Lines 396-415, and a new Figure (Figure 6):

"An additional numerical experiment was also undertaken to attempt to match the observed data to a single domain model which included resting phases, since no analytical solution is known for such a simulation. This was accomplished using COMSOL Multiphysics with identical settings to the dual domain models described above, but with a disabled immobile domain. Calibrating to the δD breakthough data recorded for the core from 9.52 m depth BGL by just varying dispersivity, but using the measured porosity, it is impossible to achieve a better fit than a NRMSE of 46%, even with an unrealistically high dispersivity. A better fit is possible (NRMSE = 9%, NSMEC = 0.9) if porosity is decreased to 0.1 but, again, only with an unrealistically high value for dispersivity of $1000L_c$, see Figure 6. While such a model may be useful to suggest that the effective porosity of the core through which solute is moving is much less than the total porosity, it is only possible to fit the early time data (e.g. only the first flow stage) very accurately, at the expense of the later time data. Perhaps more

importantly than the lower NSMEC (or higher NRMSE) compared to the dual domain models, the single domain model also misses a key feature of the observed breakthrough curves, the decrease in concentration during resting phases. Instead, modelled concentrations increase during resting phases as would be expected in a single domain model due to redistribution of the solute along the core by diffusion. This additional numerical experiment thus strengthens the conclusions of the study, that dual domain behaviour is indicated by our interrupted flow experiment observations, and that single domain models are inappropriate as a means of analysis.

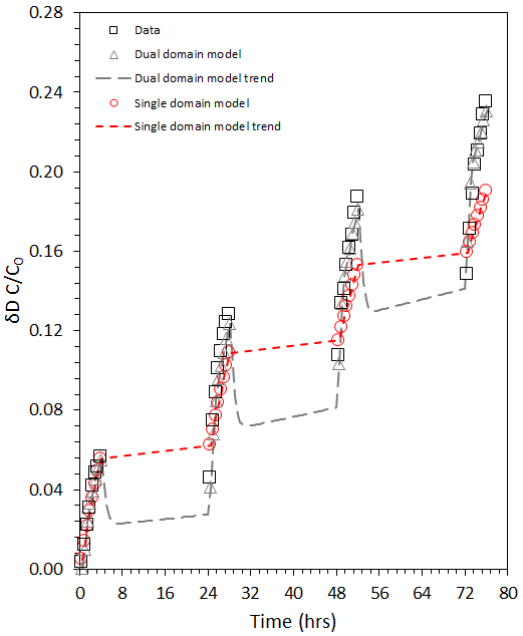

Figure 6. Comparison of single and dual domain interrupted flow transport model output for the core taken from 9.52 m BGL."

With regard to the issue about the rising limb of the breakthrough curve, we understood the reviewer's initial comment to mean that we hadn't included the resting phases in our model and sort to clarify in our original reply that we had indeed modelled the whole breakthrough curve, including resting phases. We see from the additional comment that the reviewer is actually asking about data from the resting phases but since no effluent is produced during the rest phases (by definition, since there is no flow during these periods), there are no data points that can be collected between the stopping and starting of flow.

Reviewer #2

The reviewers' comments were carefully taken into account and the manuscript was properly modified, improving the overall quality and significance of the paper. Therefore, I recommend this manuscript for publication, after some minor technical corrections listed below.

- Line 263: in the rest of the paper the symbol ϕ, and not ϕ_T, has been used to denote the total porosity.

We have removed the subscript T.

- The core length has been denoted sometimes with L (lines 272, 276, 278, 323, 374, Table 3) and sometimes with L_C (line 148, Tables 1 and 2).

We have added subscript c to all core length notation.