Responses to referee comments for “Technical Note: The use of an interrupted-flow centrifugation method to characterise preferential flow in low permeability media” (hess-2014-518) by R. A. Crane et al.

Many thanks to Anonymous Referee #2 for giving their time to provide comments on our manuscript. Please see below for our responses to your questions/comments.

Anonymous Referee #2

1 General comments

The manuscript proposes a novel method to characterise preferential flow in low permeability media with preferential flow paths. The method is given by a combination of an experimental and a modelling phase. The experimental phase consists in performing laboratory transport tests using the interrupted-flow centrifugation method. The modelling phase consists in fitting the experimental results with a dual domain model which is a variant of the dual porosity model considering also the molecular diffusion in the immobile domain. The method is applied to three samples of smectite clay dominated samples, with diameter around 10 cm and height between 3 and 5 cm. The results show that the dual domain model fits very well the experimental data and that the model fitting efficiency is relatively insensitive to the model parameters. The conclusion is that the method proposed is a powerful method to characterise preferential flow in low permeability media. I think that the subject is of interest to the scientific community: as stated by the authors, the hydraulic properties of aquitards are important, as the presence of preferential flow paths can compromise their integrity as barriers to the movement of groundwater contaminants. The method proposed in this paper has the objective of characterizing the dual porosity behaviour of such porous media. The article is well structured, well written, concise and generally clear. The objectives are clearly stated both in the abstract and in the introduction, where a good list of references is given to introduce the subject. Therefore, I recommend this manuscript for publication, after some minor revisions suggested in the following.

I have some doubts about the applicability of the method to practical situations. Specifically:

• I wonder at which scale this method can be applied. The samples diameter is around 10 cm and their height between 3 and 5 cm. Can the best fit parameters obtained at this scale be used to model transport at larger scale?

This is a good question, but beyond the scope of this paper – it can only be effectively answered, with the integration of the results from this work with well constrained field data, which is something we are now working on for a future paper. However, as we note in the current paper, a key benefit of the use of a relatively large centrifuge permeameter (100 mm diameter) for this method is their ability to house large samples compared to conventional core testing (~20 to 30 mm diameter). This has provided a unique capability to identify dual porosity at a scale of centimeters that would be difficult or impossible to quantify at other scales.

• Does the centrifugation affect the structure of the porous medium, i.e., the connectivity of the preferential flow paths?

Whether or not there is significant alteration of porous structure depends upon a wide range of complex physical and chemical factors associated with the core and the centrifuge induced stress environment. In this work, in order to minimize any unnatural physical effects such as new or enhanced preferential flow paths, the maximum centrifuge g-level was set at slightly less than the estimated in situ stress of the core sample at the depth from which it was extracted. Maximum stress to prevent deformation of the core was estimated assuming that the overlaying formations were fully saturated and of the same density to the core samples, using Equations 3 and 4 (see Section 2.2.). In addition, this centrifuge permeameter was designed to position at a relatively large centrifuge radius of 0.65 m to reduce stress differences (due to g-level) between the base and top of the core, compared with small centrifuge permeameters.

• One of the conclusions of the paper is that the modelling enabled aspects of the physical properties of the two domains to be inferred. Moreover, the model fitting is shown to be relatively insensitive to the model parameters; can this fact weaken the former conclusion?

The numerical modeling was used as a complimentary technique to confirm whether the shape of curve was indicative of dual porosity behavior. The parameter with the greatest sensitivity was the mass transfer coefficient which is also the parameter which lends greatest insight into the likely geometry of the preferential flow paths. So, in that sense, we consider the conclusion that aspects of the physical properties of the two domains can be inferred from the model is sound. However, we have noted in the text that further work is needed to clarify the relationship between the model parameters and the physical structure of the cores by other methods.

Page 70, line 23: I suggest to modify the sentence in ’the possible dual porosity behaviour’ or in ’low permeability porous media characterised by the presence of preferential flow path’. In fact, if the low permeability porous medium is homogeneous, then it does not show any dual porosity behaviour.

Yes, we agree and have now added the word ‘possible’ before dual porosity as the reviewer suggests.

• Page 72, line 3: I think it should be specified somewhere in this section that three core samples were analysed, each one coming from a different depth.

We agree and have added an additional sentence in Page 23 Line 4:

In total three core samples were analysed, which were taken from depths of 5.03, 9.52 and 21.75 m BGL.

• Page 72, lines 16-19: Is there a reason why the clay cores are taken larger than the core holder and then trimmed, instead of taking directly a clay core of the needed diameter? I think some more explanations would be useful here. Moreover, I think the sentence should be rephrased, as it seems that the subject of the verb 'inserted' is 'the outer 5 mm of the clay cores'.

We agree and have now added the following sentence to Line 19:

The cores were trimmed in order to remove any physical and chemical disturbance associated with the core extraction (drilling) process and to ensure a close fit with the permeameter liner.

We have also changed Lines 16-19 to: Prior to mounting into the CP the outer 1.6 mm of the clay cores were trimmed and the trimmed cores were then inserted into Teflon cylindrical core holders (100 mm internal diameter, 220 mm length) using a custom built mechanical cutting and loading device.

This makes it clear that the trimmed cores were inserted into the core holders rather than the trimmed material.

• Page 74, line 20 (and following): I think it should be clearly distinguished between the different dual domain approaches in order to correctly situate the model introduced by the author in the existing literature. In particular, Coats and Smith (1964) and van Genuchten and Wierenga (1976) introduce a model of mobile/immobile type (which I would call dual porosity model). On the other hand, the model introduced by Gerke and van Genuchten (1993, 1993a) is not a mobile/immobile model, as water can flow in both domains (with different velocities) and so the solute can be transported by advection and by dispersion in both domains. This kind of models is more correctly called 'dual permeability model' (see, e.g., Baratelli et al 2014 for more references to the two different modeling approaches). The model introduced by the authors is closer to the dual-porosity mobile/immobile approach.

We agree that it is important to be clear that the model presented is a "dual porosity" model and not a "dual permeability" model. We have therefore removed the reference to Gerke & van Genuchten (1993) at this point in the manuscript which is presumably the source of the reviewer's concern since, as they correctly point out, it refers to a dual permeability approach.

• Page 76, equations (5)-(7): I think the authors should clearly state how their model is ’novel’ (Page 70, line 23) with respect to the model already existing in the literature. I guess the novelty is mainly related to the presence of the molecular diffusion term in the immobile domain; it would be interesting to add some explications to justify this choice.

We stated in the text (p.76, lines 3-7) that the main novelty in the model formulation which is the setup of the upper boundary condition switching during the flow and rest phases respectively. Hence, we’re not sure what the reviewer’s concern is here. However we have clarified the boundary conditions as described in our reply to the next comment by the reviewer “Page 76, lines 1-9: I think that the boundary conditions used are not very clear.”

• Page 76, lines 1-9: I think that the boundary conditions used are not very clear. In particular, the flow is not simulated and so the boundary conditions for flow (line 1) are not required. Moreover, it would be useful to explain more clearly the boundary conditions applied at the top and bottom of the columnn for both the mobile and immobile domain.

Yes, we agree this section needs clarification and the mention of a flow boundary condition was misleading (we had explicitly included the flow equations in an earlier iteration, but not for the results finally presented here). We have now clarified this section (originally p76, lines 1-9) to read:

The initial concentration conditions were set to zero for both domains for all model runs. During centrifugation periods, a variable solute flux upper boundary condition was used for the mobile domain varied according to the product of the measured fluid flux & input concentration (C_0) during each experiment as follows:

$$\frac{q(t)}{\emptyset_m}C_0 = \frac{q(t)}{\emptyset_m}C_m + D_m\frac{\partial C_m}{\partial z}$$

A Dirichlet (constant concentration) upper boundary condition was used for the immobile domain during times of centrifugation. A novel aspect of the models, facilitated by the flexibility of model structure variations possible in COMSOL, was that the upstream transport boundary for both domains was switched to a zero flux condition during the interrupted flow phases. The downstream transport boundary conditions for both domains were given by:

$$\frac{\partial C_{m,im}}{\partial z} = 0$$

At $z = L$, where L was sufficiently large to ensure the results at the column outlet distance (at $z \ll L$) were not sensitive to the position of the boundary.

• Page 78, line 22-23: It would be interesting to explain the implications of this result.

We have attempted to do this in the text on p.78 lines 24-28.

• Page 79, line 3-4: Is the choice of using the same _ as for parallel fracture geometry justified?

Good question! In order to show the range of uncertainty associated with this geometry factor we have now included a range of inferred values of *a* using Beta values for parallel slabs (Beta = 3) as well spherical aggregates (Beta =15).

3 Technical corrections

• Page 72, line 7: Correct ASTM 2012 with ASTM (2012).

We have now corrected this.

• Page 72, lines 11-12: I suggest to explicitly define the symbols EC and Eh, although I understand that it is a rather standard notation.

We have now defined these symbols, see tracked changes.

• Page 72, equation (2): It seems to me that the results has the unit of [1/T] and not of [L/T] as I would expect for the hydraulic conductivity.

Equation 2 gives dimensions of K of [L/T], following the detailed derivation provided in ASTM 2000 and Timms et al. (2014). Equation 2 is expressed for laboratory convenience in terms of RPM rather than more standard parameters, Dimensions for $(RPM)^2$ of [1/L] is obtained, such as equation 5 of Timms et al. (2014), by rearranging $a/g = 0.001.r.(RPM)^2$. The [1/L] dimensions of RPM^2 then cancel with the other dimensions in Equation 2 of this paper to give K with dimensions of [L/T] as expected.

• Page 72, equation (2): I think K should be substituted by Kv, as in the following the hydraulic conductivity is always indicated as Kv without being explicitly defined (see page 78, Table 1, ...).

We have now changed this.

• Page 76, line 2: exchange -> exchanged.

We have now changed this.

• Page 76, line 28: I think that _ should be corrected with _T .

The notation we have used for the total porosity here is consistent with how we defined it on p75 line 10, so we don’t think it needs changing.

• Page 78, line 19: I suggest to add a comma between ’0.43’ and ’this’.

We have now changed this.

• Page 82, line 21: afield -> a field.

We have now changed this.