

**To Prof. E. Zehe**  
**Editor, Hydrology and Earth System Sciences (HESS)**

**Feedback mechanisms between precipitation and dissolution reactions across  
randomly heterogeneous conductivity fields**

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Dear Editor,

We appreciate all efforts invested by you and the Reviewer in our manuscript and are grateful for the opportunity to resubmit a revised draft for your consideration. We address all comments from the Reviewers in the following itemized lists, which we arrange enclosing each answer within corresponding review items. Please, note that Reviewer's comments and our responses are given in black and blue fonts, respectively.

Sincerely,

Yaniv Edery, Martin Stolar, Giovanni Porta, Alberto Guadagnini

**Replies to Reviewer #1**

The paper addresses a very timely and relevant topic in reactive transport through heterogeneous porous media. The most significant and novel result concerns the significant changes in non-Fickian transport behaviors while the while velocity variance display modest variations.

[We thank the Reviewer for their careful review and positive comments.](#)

The topic is suitable for HESS and I recommend minor revisions related to:

- The chemical model is a very simplified description of dissolution/precipitation processes. It should be better justified through additional references (see references below) and some quantitative information should be provided to describe its field of application. Without this information, the paper may appear like discussion around numerical experiments. Furthermore, the 'strange' dimension of the domain (60 x 24 cm<sup>2</sup>) seems related to a lab. scale experiment....

[While simplified, the chemical set-up follows previous work by Edery et al. \(2011\), where there is an extensive assessment of the employed formulation. This analysis is also summarized in the supplementary material in this study. Our revised manuscript now reads \(lines 104-106\): "While simplified \(see, e.g., Lichtner, 1988; Dreybrodt et al., 1996\), the chemical set-up follows previous work by Edery et al. \(2011\), where there is an extensive assessment of the employed formulation \(see also the Supplementary Material\).](#)

[The Reviewer is right. In line with the above mentioned work, our study is representative of a laboratory scale analysis. Yet, the precise dimensions have no particular implication and they are only selected to ensure a meaningful description of the correlation structure is included](#)

in the initial conductivity field. Our revised manuscript now reads (lines 133-136): “We note that, while our study is representative of a laboratory scale analysis, the dimensions of the domain have no particular implication and they are only selected to ensure a meaningful description of the correlation structure that is included in the initially generated conductivity fields.”

## References

Y. Edery, H. Scher and B. Berkowitz, *Water Resources Research* **47** (8) (2011).

- A key parameter in dissolution/precipitation is the surface at the interface between solid and fluid, which is insufficiently discussed in the paper. Porosity and hydraulic conductivity are modified by precipitation/dissolution. What about the area in contact with the fluid?

We agree with the Reviewer that surface area is a key parameter when a kinetic precipitation-dissolution model is considered. Here, we consider local equilibrium. The precipitation and dissolution terms directly descend from algebraic calculations related to the local fluid equilibrium and are therefore not related to surface area. Note that this choice is functional to the specific objective of this work which is related to the characterization of transport under dynamic evolution rather than a detailed characterization of the reactive process and its effect on the pore-scale structure.

Our revised manuscript now reads (lines 214-219): “We also recall that we consider local equilibrium, this choice being consistent with the specific objective of this work which is related to the characterization of transport under dynamic evolution of continuum-scale quantities such as conductivity, rather than being focused on a detailed characterization of the effects of the reactive process on the pore-scale structure (as reflected, e.g., by local changes of specific surface area).

- 20 random realizations is a limited number of realizations... Can you discuss this number a posteriori, i.e. by analyzing the variability within realizations?

We have analyzed convergence of the results with respect to the number of realizations. As an example of the type of results we obtain, Figure R1.1 depicts the average of the temporal evolution of the sum of all conductivity changes  $|\Delta K|$  in the domain as a function of the number of realizations considered for the most heterogeneous setting (values of conductivity changes being evaluated across temporal windows of width equal to 1 minute, as in Figure 4b in the manuscript). These results show that the number of realizations we employ is sufficient to characterize the reactive process, as quantified through the metric depicted. Results of similar quality are obtained also for the remaining values of initial log-conductivity variance examined.

While we think including Figure R1.1 in the revised manuscript will not bring additional value to the study, we explicitly refer to such a preliminary analysis in our revised text (see lines 198-207 of our revised manuscript).

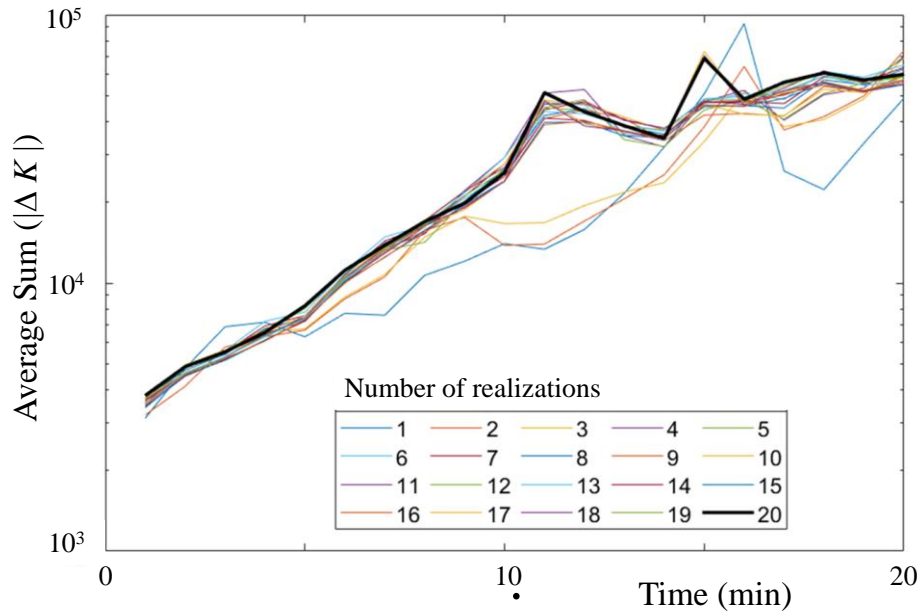


Figure R1.1. Temporal evolution of the sum of all conductivity changes  $|\Delta K|$  averaged according to an increasing number of Monte Carlo realizations for the setting associated with initial log-conductivity variance  $\sigma_0^2 = 5$ . Values of conductivity changes are evaluated across temporal windows of width equal to 1 minute, as in Figure 4b in the manuscript.

- The particles are injected at regular time interval. Do they all start at the same time or is the injection time uniformly distributed over the time step?

Particles are injected at the very beginning of each time step (each time step is of 6 s). Our revised manuscript now reads (lines 150-151): “Particles are injected at the very beginning of each time step  $\Delta t$  and ...”.

- I do not understand why there is the number 3 in formula line 153.

We fixed the typo and we thank the Reviewer for alerting us.

- I assume that the 20 minutes are for the initial pore volume (L234) ?

Indeed, the Reviewer is correct and this is clarified in the revised manuscript (lines 272-274): “Figure 3a and b depict a heat map of the  $h$  particles distribution at time  $t = 20$  min (i.e., corresponding to the first pore volume), ...”.

#### References:

Lichtner, The quasi-stationary state approximation to coupled mass transport and fluid-rock interaction in a porous medium, GCA, 1988.

Dreybrodt et al., The kinetics of the reaction  $\text{CO}_2 + \text{H}_2\text{O} + \text{H}^+ + \text{HCO}_3^-$ , as one of the rate limiting steps for the dissolution of calcite in the system ..., GCA, 1996.

We thank the Reviewer and included these references in our revised manuscript.

## Replies to Reviewer #2

The manuscript, by Edery et al., addresses a significant topic of reactive transport in heterogeneous porous media, with precipitation/dissolution scenario. The main contributions of this paper are variation/changes in the spatial-statistical moments of the velocity field during the transport of reactive species, and how different flow zones (i.e., preferential paths and low conductivity zones) are contributing (differently) to precipitation/dissolution.

The manuscript deals with a timely and relevant topic, well-written and presented, and its results have a general contribution for various systems in hydrology and earth science. Therefore, it is suitable for publication in HESS, with a few minor revisions.

We thank the Reviewer for their positive appraisal of our study and the time invested in reviewing our work.

General comments:

- The authors use a Darcy scale model to solve the velocity field, and thus the transport, while the system is “only” 60\*24 cm (Lab scale). More important, for Darcy scale, I would expect the use of dispersion coefficient and not diffusion (Eq. 4). Although understanding the authors choice here, in order to use particle tracking approach, they should discuss their choice (i.e., diffusion over dispersion), and emphasize the relevance of their results for larger scale systems (adding references).

We thank the Reviewer for addressing this important point. The scale of the system is indeed intended to mimic a laboratory scale scenario. Our modeling choice is motivated by the reasons detailed in the following. In general, we acknowledge that using a velocity-dependent dispersion would be an option, enabling one to take into account pore-scale processes within a continuum-scale model. It is our view that this would add an additional level of complexity to our numerical simulation without modifying the key elements of our work, which is focused on the interaction between flow patterns and reactive processes. In this context, our choice is to neglect local dispersion while approximating the effect of fluctuations of velocity arising between diverse realizations of the conductivity field. Averaging in a multi-realization context enables us to represent such effects, which are commonly termed as macrodispersive effects. In line with this reasoning, our choice is to rely on a constant and isotropic diffusion coefficient that can be associated with an advection-dominated transport regime (as quantified in terms of a Péclet number, see below). This choice is also consistent with previous works (e.g., Aquino and Bolster, 2017; Wright et al., 2021)

Our revised work now reads (lines 166-175): “It is noted that, while taking into account pore-scale processes within a continuum-scale model through a local scale dispersion could be a modeling option, this would add an additional level of complexity to our numerical simulation without modifying the key elements of our work, which is focused on the interaction between flow patterns and reactive processes. In this context, our modeling choice is to represent macrodispersive effects through averaging (in a multi-realization context) the effect of fluctuations of velocity arising between diverse realizations of the conductivity field. Consistent with this, we rely on a constant and isotropic diffusion coefficient that can be associated with an advection-dominated transport regime (as quantified in terms of a Péclet number, as seen in the following). This choice is also consistent with previous works (e.g., Aquino and Bolster, 2017; Wright et al., 2021).”

**References**

E. E. Wright, N. L. Sund, D. H. Richter, G. M. Porta and D. Bolster, *Stochastic Environmental Research and Risk Assessment*, 1-19 (2021).

T. Aquino and D. Bolster, *Transport in Porous Media* **119** (2), 391-402 (2017)

- The chemical model (sec. 2.1). I am not sure that “infinite Damkholer number” (line 98) is the correct definition. How much the reaction products are sensitive to the choice of numerical parameters (time interval for reaction), overall particles and the grid size?

We note that the expression infinite (local) Damköhler is related to the observation that reactive processes in our study take place under the assumption of instantaneous local equilibrium. The  $Da$  number usually provides an appraisal of the ratio between advective (or diffusive) and reactive time scales. The reactive time scale approaches zero at equilibrium. In this sense, we can state that  $Da \rightarrow \infty$ . We clarify this point by stating that (see lines 98-100 of the revised manuscript): ... (i.e., as the reactive time scale approaches zero at equilibrium, this scenario corresponds to a local Damköhler number which tends to infinity) ...”.

Regarding the sensitivity of the results to the numerical parameters, we have run several tests. With reference to the sensitivity of the results to the numerical parameters, we note that: (a) when considering single realizations, the results showed only minute sensitivity to increasing the number of particles by a factor of 10 (less than 3% difference in the results was observed); and (b) results display a slightly larger sensitivity to the time step, otherwise resulting in a difference of less than about 5% in the amount of reaction when decreasing the time interval by an order of magnitude.

We had performed a test to assess possible influences of the grid size on the key results of the study. We did so by considering a numerical grid with elements characterized by half the size of those included in the manuscript. Due to constraints related to computational time, we performed this analysis for 10 random realizations of the heterogeneous conductivity fields associated with the highest initial variance (i.e., variance equal to 5) and comprising  $120 \times 120$  and  $240 \times 240$  elements of uniform size  $\Delta = 0.1$  and  $0.2$  cm, respectively (the value  $\Delta = 0.2$  cm being then used in the manuscript).

As an example of the type of results we obtain, Figure R2.1 depicts the average across the considered 10 realizations of the temporal evolution of the sum of all conductivity changes  $|\Delta K|$  in the domain taking place, values of conductivity changes being evaluated across temporal windows of width equal to 1 minute (see Figure 4b in the manuscript). These results show that the overall trend is not significantly affected by the grid size. Results of similar quality are obtained also for the remaining values of initial log-conductivity variance examined. This analysis imbued us with confidence about the quality of the results obtained with the grid size employed in the manuscript, which we selected as good compromise between computational accuracy and execution time constraints, in light of the objectives of the study.

Our revised text now states: (see lines 198-207 of the revised manuscript): “With reference to the sensitivity of the results to the numerical parameters, we note that: (a) when considering single realizations, the results showed only minute sensitivity to increasing the number of particles by a factor of 10 (less than 3% difference in the results was observed); (b) results display a slightly larger sensitivity to the time step, otherwise resulting in a difference of less than about 5% in the amount of reaction when decreasing the time interval by an order of magnitude; and (c) preliminary analyses aimed at assessing possible influences of the grid size on the key results of the study imbued us with confidence about the quality of the results obtained with the grid size employed in the study (details not shown), which we select as good compromise between computational accuracy and execution time constraints, in light of our objectives.”

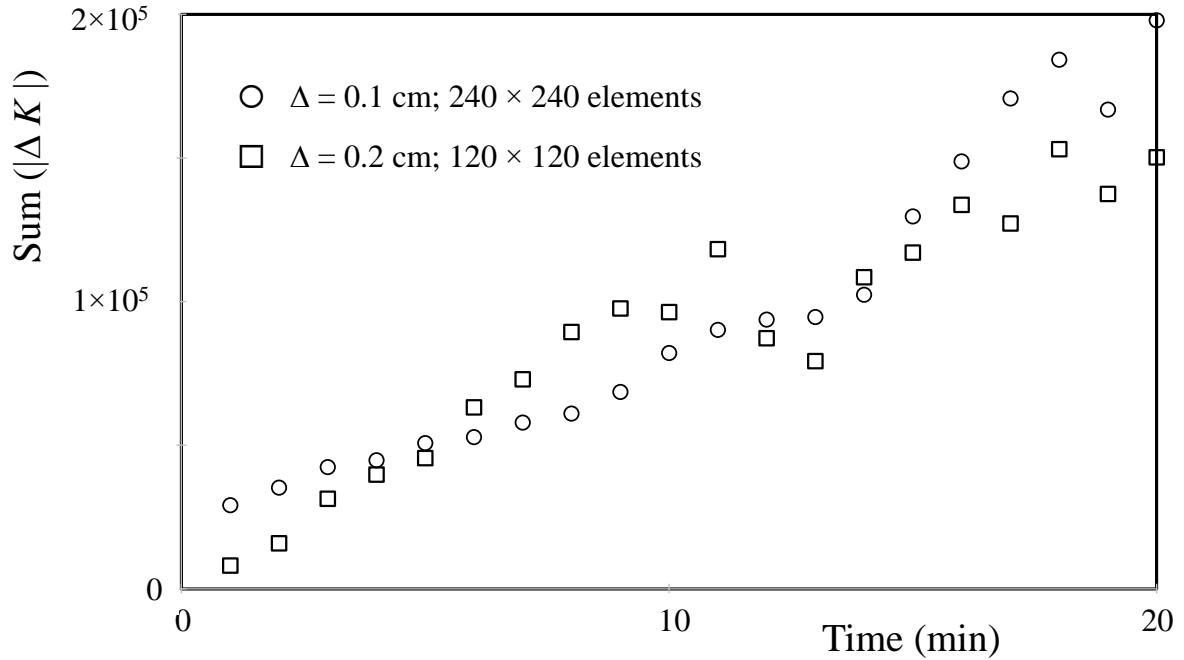


Figure R2.1. Temporal evolution of the sum of all conductivity changes  $|\Delta K|$  (averaged across 10 realization) for the setting corresponding to initial log-conductivity variance  $\sigma_0^2 = 5$ . Values of conductivity changes are evaluated across temporal windows of width equal to 1 minute, as in Figure 4b in the manuscript. Results are depicted for grids comprising  $120 \times 120$  and  $240 \times 240$  elements of uniform size  $\Delta = 0.1$  and  $0.2$  cm, respectively (the value  $\Delta = 0.2$  cm being then used in the manuscript)

- The relation between the non-Fickian transport behavior and the CTRW pdf TPL. Although the fits of  $\psi(t)$  are reasonable (Fig. 2), the authors might add a fit to the breakthrough curve, for a selected realization, to show that the transport in a non-reactive system follows the ctrw model (TPL). Also, how the authors find the TPL parameters (table. 1)? Are they related to the statistical moments (i.e., particle mean velocity and variance)?

A major finding of this study is documenting that spatial heterogeneity of reaction leads to (a) the emergence of non-Fickian transport in settings where a Fickian model can be used to characterize non-reactive solute migration prior to reaction (i.e., the scenario corresponding to a unit log-conductivity variance of the original system); or (b) enhancing the non-Fickian pattern associated with non-reactive solute migration prior to reaction (i.e., the scenarios corresponding to the high log-conductivity variances of the original system).

Quantification of departure from a Fickian pattern is here addressed through the tail of the probability density function (PDF) of particle waiting times for a tracer test performed on the conductivity field prior and after reaction has taken place.

In this context, we do not explore the skill of a model such as, e.g., the CTRW (mentioned by the Reviewer), to interpret non-reactive transport across the conductivity fields prior and/or after reaction is documented. We note that some of the parameters of a CTRW model can be estimated directly from the above mentioned PDF. Namely, the slope of the tail of the PDF and the values of  $t_1$  and  $t_2$  can be estimated through a log-log representation of the

type depicted in Figure 2 of the original manuscript. We will add this clarification in the revised manuscript.

Given our objective, we did not fit breakthrough curves (BTCs) associated with the non-reactive transport simulations in our study, a feature which is then out of the scope of our study.

We note that fitting a BTC with a CTRW model formulation would require estimating the dispersion coefficient, while relying on the parameters estimated through the above mentioned PDF and the arithmetic mean velocity in the domain. This would render an effective one-dimensional CTRW-based representation of the BTC of the kind that is typically presented.

Following the request the Reviewer, we employ a one-dimensional CTRW model to interpret BTCs associated with non-reactive transport taking place in the original conductivity fields and at various times after reaction takes place while (a) accounting for the parameters of the PDF of the of particle waiting times and mean velocity and (b) fitting dispersion.

Table R.2.1 lists the values of the parameters obtained from the PDF of the of particle waiting times (i.e.,  $\beta$ ,  $t_1$ , and  $t_2$ ) evaluated as described above, of the mean velocity ( $\bar{v}_T$ ), and of the dispersion coefficient ( $D$ ), the latter being obtained through a least-square fit of a one-dimensional CTRW model against numerical BTCs, for selected single realizations of log-conductivity fields with initial variance  $\sigma_0^2 = 1$  and 5 and some observation times ( $t = 0$  corresponding to the conductivity distribution prior to reaction).

One can clearly note the increasingly non-Fickian nature of transport from the values of the parameters obtained from the PDF of the of particle waiting times (i.e.,  $\beta$ ,  $t_1$ , and  $t_2$ ). This corresponds to the major finding associated with our study and does not require relying on a CTRW transport formulation.

	$\sigma_0^2 = 1$			$\sigma_0^2 = 5$		
$t$ [min]	0	10	20	0	10	20
$D$	0.007	0.085	0.91	0.27	0.345	0.372
$\bar{v}_T$	0.36	0.37	0.38	0.45	0.46	0.47
$\beta$	2	1.4	1.05	1.4	0.8	0.6
$t_1$	0.1	0.09	0.06	0.08	0.03	0.01
$\text{Log}_{10}(t_2)$	1	2.5	2.8	1.6	2.5	3

Table R.2.1. Values of the parameters obtained from the PDF of the of particle waiting times (i.e.,  $\beta$ ,  $t_1$ , and  $t_2$ ), of the mean velocity ( $\bar{v}_T$ ), and of the dispersion coefficient ( $D$ ), the latter being obtained through a least-square fit of a one-dimensional CTRW model against numerical BTCs, for selected single realizations of log-conductivity fields with initial variance  $\sigma_0^2 = 1$  and 5. Results corresponding to some selected observation times (prior and after the reaction takes place) are listed.

Otherwise, Figure R2.2 depicts the results obtained through the one-dimensional CTRW formulation associated with the parameters listed in Table R.2.1 and the BTCs evaluated in our numerical analyses. We stress that, as stated above, these are obtained upon estimating dispersion through a CTRW fit.

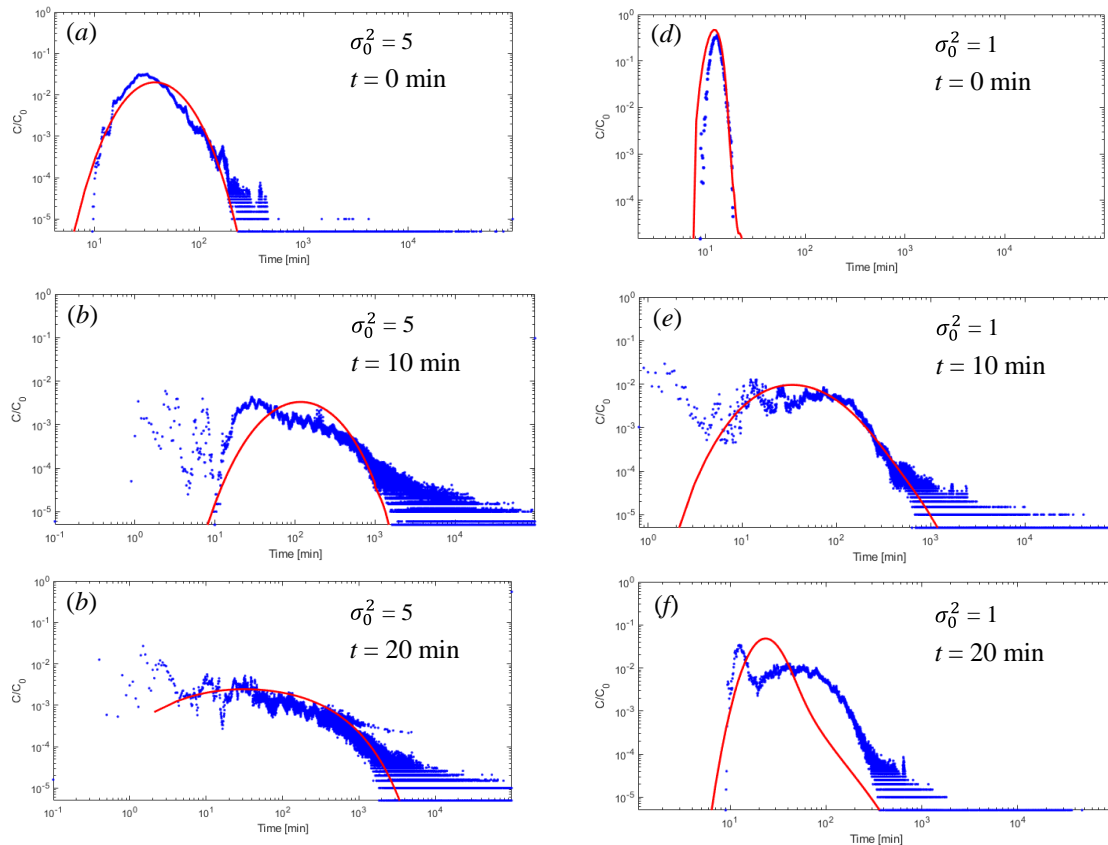


Figure R2.2. Results obtained through the one-dimensional CTRW formulation associated with the parameters listed in Table R.2.1 and the BTCs evaluated in our numerical analyses.

These latter results clearly show that the non-Fickian nature of transport, as embedded in the parameters characterizing the PDF of the of particle waiting times (i.e.,  $\beta$ ,  $t_1$ , and  $t_2$ ), is not captured entirely by the 1D solution of the CTRW transport formulation when considering the log-conductivity fields resulting from reaction. As we state above, our study is not aimed at exploring the skill of a model such as, e.g., the CTRW, to interpret non-reactive transport across the conductivity fields prior and/or after reaction takes place and we will defer this particular analysis to a subsequent study.

We clarify these elements in our revised Conclusions where we now state (see lines 424-429 of the revised manuscript): “Solute mass weighted conductivity and velocity distributions are at the basis of our characterization of the parameters of a TPL model which is at the core of the characterization of the probability density function of particle travel/waiting times and enables us to capture effective (upscaled) non-Fickian transport behaviors. With the progress of precipitation/dissolution reactions, transport shifts towards an increasingly acute non-Fickian effective behavior (see Figure 2 and ensuing parameter values listed in Table 1).”

We then state (see lines 437-440 of the revised manuscript): “In this context, we recall that our study is not aimed at exploring the skill of any specific transport model to interpret non-reactive transport across the conductivity fields prior and/or after reaction takes place, this particular analysis being deferred to a future study.”



In this context, maybe to add the mean velocity value to table. 1.

Values of mean velocity and of the weighted velocity for the tracer test are now included in the revised Table1.

- What is the magnitude of  $Pe$ . Seems that the transport is at (relatively) high  $Pe$  (advective dominant), and thus the occurrence of preferential paths is more relevant. However, many water resources are usually under low  $Pe$  value.

We consider the following formulation for the Péclet number,  $Pe$  (see, e.g., Huysmans and Dassargues, 2005):

$$Pe = \frac{V_d \sqrt{k}}{n_e D_d} = \frac{V_d \sqrt{K \frac{\mu}{\rho g}}}{n_e D_d} = \frac{5 \cdot \sqrt{1 \cdot \frac{0.0105}{10^5}}}{0.3 \cdot 10^{-5}} = 540$$

where  $V_d$  is Darcy velocity;  $k$  is the intrinsic permeability;  $n_e$  is effective porosity;  $K$  is (arithmetic) average;  $\mu$  and  $\rho$  are dynamic viscosity and density of water, respectively;  $g$  is gravity; and  $D_d$  is diffusion. The ensuing value of  $Pe$  is indeed indicative of an advection-dominated transport setup. An additional study on the sensitivity of the results to the magnitude of  $Pe$  for a completely uniform conductivity field is currently under completion and preliminary results confirm that the same feedback mechanism between reaction and evolution of preferential pathways is observed. We do agree that results might be different in a diffusion-dominated system and would require additional analyses. We are planning of tackling these in a separate study because the simulations will require a more efficient computational methods for the implementation of the reaction and transport.

## References.

M. Huysmans and A. Dassargues, *Hydrogeology Journal* **13** (5-6), 895-904 (2005)

- The system seems to be sensitive to the choice of the boundary conditions, where the low velocity sampling (which contributes most to the non-Fickian behavior) is only around the inlet boundary. The authors should discuss that in more details and add more references (e.g., Kang et al., 2020 <https://doi.org/10.1029/2020WR027378>)

We start by thanking the Reviewer because their comment enabled us to clarify that the original manuscript included an inappropriate description of the meaning of the diverse colors employed to illustrate the results depicted in Figure 3g&h. The green color in Figures 3g&h marks the **decrease** in conductivity (and not the **increase** in conductivity as written in the original version of the manuscript), this being also consistent with the color palette in Figure 3e&f. The red color in Figures 3g&h marks the **increase** in conductivity and not the **decrease** in conductivity, this being also consistent with the color palette in Figure 3c&d. These elements are now fixed in the revised manuscript.

The documented conductivity increase near the inlet is consistent with the constant low pH value introduced by the step function of particles injected in the system which leads to a high dissolution rate and therefore higher conductivity near the inlet, an effect evidenced in several studies (Fredd and Fogler, 1998; Nogues et al., 2013; Kang et al., 2013; Molins et al., 2014; Jung and Navarre-Stichler, 2018). The study suggested by the Reviewer (Kang et al., 2020) deals with fractured rocks and the inlet effect therein is regarded to be the result of dead-end fractures, which is not the case in our study. We agree that the initial/boundary conditions can affect non-Fickian transport features, as it has been demonstrated in a number of works

also referring to flow in porous media simulated via Darcy scale models. The injection mode will certainly affect solute transport close to the inlet boundary, which will then be reflected in the recorded residence times and the related statistics. We prefer to adhere here to the investigated set of boundary conditions (flux-weighted injection, mean uniform flow), which are representative of a well-studied framework and not to delve into a discussion on this point. We feel the latter would only distract the reader from the key messages of the study.

The point is mentioned in the revised Conclusions which now read (see lines 452-455 of the revised manuscript): “Our results are based on numerical simulations and may be used to inform upscaling approaches to capture the pre-asymptotic and asymptotic dynamics of reactive transport in heterogeneous systems through simplified models. Future computational studies might also include an assessment of the importance of initial/boundary conditions and/or of solute injection mode on the emergence of non-Fickian transport features, with special emphasis on locations close to the inlet boundary which might then have an impact on the overall solute residence times and related statistics.”

### References.

- C. N. Fredd and H. S. Fogler, *AICHE journal* **44** (9), 1933-1949 (1998).  
Q. Kang, D. Zhang and S. Chen, *Journal of Geophysical Research: Solid Earth* **108** (B10) (2003).  
J. P. Nogue, J. P. Fitts, M. A. Celia and C. A. Peters, *Water Resources Research* **49** (9), 6006-6021 (2013).  
S. Molins, D. Trebotich, L. Yang, J. B. Ajo-Franklin, T. J. Ligocki, C. Shen and C. I. Steefel, *Environmental science & technology* **48** (13), 7453-7460 (2014).  
H. Jung and A. Navarre-Sitchler, *Geochimica et Cosmochimica Acta* **234**, 70-83 (2018).

### Specific comments:

- In the Kozeny-Carmen model, the authors might add the formula for the porosity evolution.

Done (see (5) in the revised manuscript).

- Lines 171-172: what will be the effect of different boundary constrains?

This point is clarified in the revised manuscript (see lines 190-193) which now reads: “These constraints are set for consistency with our assumptions, i.e., to consider Darcy flow across the porous domain. A complete clogging (or opening) of a void space would require a different mathematical and conceptual treatment, which is beyond the scope of our study.”.

- Are 20 realizations being significant for a robust statistic? What is the variability within realizations?

We have analyzed convergence of the results with respect to the number of realizations. As an example of the type of results we obtain, Figure R2.3 depicts the average of the temporal evolution of the sum of all conductivity changes  $|\Delta K|$  in the domain as a function of the number of realizations considered for the most heterogeneous setting (values of conductivity changes being evaluated across temporal windows of width equal to 1 minute, as in Figure 4b in the manuscript). These results show that the number of realizations we employ is sufficient to characterize the reactive process, as quantified through the metric depicted. Results of similar quality are obtained also for the remaining values of initial log-conductivity variance examined.

While we think including Figure R2.3 in the revised manuscript will not bring additional value to the study, we explicitly refer to such a preliminary analysis in our revised text (see lines 198-207 of our revised manuscript).

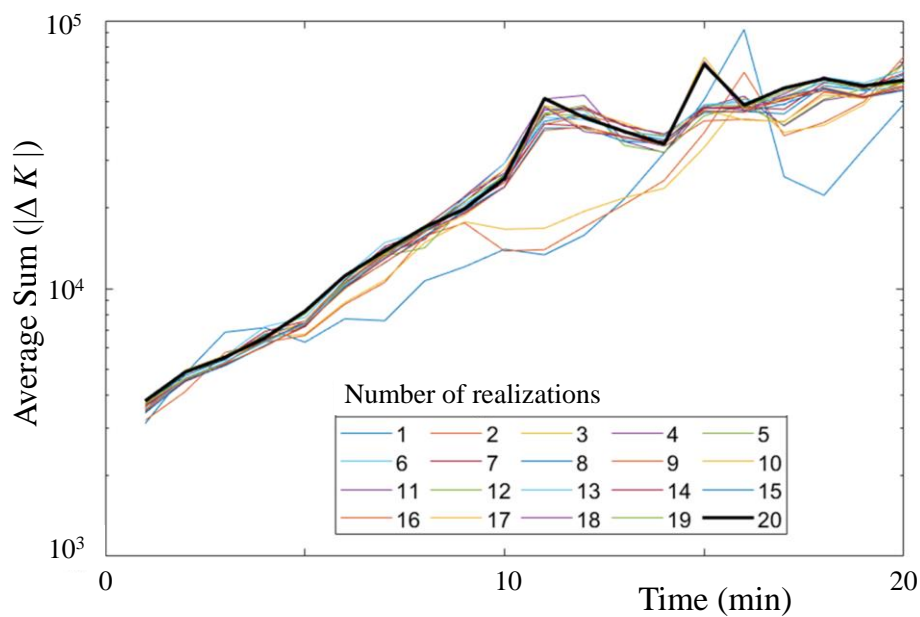


Figure R2.3. Temporal evolution of the sum of all conductivity changes  $|\Delta K|$  averaged according to an increasing number of Monte Carlo realizations for the setting associated with initial log-conductivity variance  $\sigma_0^2 = 5$ . Values of conductivity changes are evaluated across temporal windows of width equal to 1 minute, as in Figure 4b in the manuscript.

- How the pore volume is defined? By the initial condition? As the overall flux is evolving during the simulations.

Yes, the pore volume is calculated using the initial condition, i.e., before porosity and permeability are altered by the reactive processes. This is now clarified in the revised manuscript which now reads (see lines 146-147): “which is selected to represent a full pore volume (whose magnitude is evaluated through the initial condition, i.e., before porosity and permeability are altered by the reactive processes)”.

- For larger pore volumes, are the monotonic behavior in Fig. 4 might shifts? And therefore, precipitation becomes more dominant.

As pore volume increases we can obtain a higher amount of available carbonic acid due to the dissolution near the inlet, which can then be followed by precipitation downstream and hence can alter the overall conductivity and flow field. As the current study focuses on the dynamics of the first pore volume we did not address this option. Otherwise, we acknowledge the possibility to observe the result mentioned by the Reviewer. As our numerical results do not enable us to document these features, we prefer to avoid a discussion of this point in the revised manuscript.

- Line 153, please check the random walk formula for 2D

Thanks, we corrected a typo.

Technical corrections:

- Figure 3. Missing space “d and f Panels...”

Done.