Predicting the impact of spatial heterogeneity on microbial redox dynamics and nutrient cycling in the subsurface

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Response to RC3

Specific Comments:
There are a few matters that require clarification for the reader and provision of supporting information that is not in the current manuscript:

Line 170: "average hydraulic conductivity" – what average is used (arithmetic, geometric, harmonic)? I would assume arithmetic since not stated otherwise, but often the geometric mean is considered more representative of the average behavior of a heterogeneous permeability field.

The reviewer is correct in assuming that it is arithmetic average. In addition, we would like to clarify that we ensured that the average water flux in all the domains in a particular flow regime was the same by adjusting the hydraulic gradient between the inlet and outlet boundaries (L167-169).

Table 1: The "length scale" is given here as 0.1 meters, apparently as used to compute the Peclet number (ref. line 230). What is that value based on?

We apologise for the confusion. To calculate the Peclet number, the domain length was used (0.5m) to derive the Peclet number for the flow processes occurring at the observation scale. To avoid confusion, we propose to delete this entry from Table 1.

Line 195: What is the correlation length used in the simulations? I couldn't find it in the tables.

We apologise for the confusion. The correlation length referred to in this sentence is the autocorrelation spatial/length scale. Its value is 0.1 m (20% of the domain size considering that several studies have found correlation length to be 10-30% of the observation scale (Turcke and Kueper (1996) found typical correlation length to be varying from 0.16m and 0.23m in core sizes of 1.5m while Welhan and Reed (1997) observed correlation length to be 1.5 km in the total domain size of 15 km)), We will update this sentence to prevent further confusion:
" ... Each random field was characterized by the same mean value of conductivity (i.e., average conditions at the subject site (Jing et al., 2017)) and spatial autocorrelation length scale (0.1 m) in all realizations, in scaling with the size of the domain in line with previous studies (Turcke and Kuper, 1996; Welhan and Reed, 1997; Desbarats and Bachu, 1994)."

Lines 196-198: The outcomes may be sensitive to the assumption of a second-order stationary random field with horizontal anisotropy. Other types of heterogeneity (e.g., multipoint statistical models, geometric models, or depositional process models) could lead to different (and probably even more striking) conclusions. I don't view this as a flaw of the study, since this assumption is conventional, but the assumption and its potential implications should perhaps be discussed.

We agree with the reviewer that other models are available to describe spatial heterogeneity. We selected the variogram method as it condensed the representation of the spatially heterogeneous fields in a limited parameter set (de Marsily et al., 2005). Thus, it was able to represent a wide variety of heterogeneous fields regardless of geological or depositional processes involved that resulted in the creation of such spatially heterogeneous domains in the first place, or without incorporating large datasets (as required in multipoint approaches). In our work, we overcame any bias induced by the variogram approach by using the breakthrough time to indicate the extent of spatial heterogeneity, independent of how heterogeneity is described. This led us to discuss results on impact on nutrient cycling and biomass in terms of reduction in breakthrough time given the same average flow conditions. We propose that the same approach could be followed regardless of the heterogeneity model used to generate such spatial random fields. We will add a note on this aspect in the Discussion section of the revised manuscript.

Lines 245-246: Clarification is needed here to elucidate what is meant by "fit of the model" in defining the AIC criterion. It isn't clear whether this is fit to actual field observations (there are some discussed in the paper but they are not described in detail) or fit to analytical solutions (e.g. Figure S8), or something else.

We understand the source of ambiguity, we have updated this section as follows:

"

To evaluate the key factors determining the impact of spatial heterogeneity on nutrient cycling, we undertook a series of multivariate statistical analyses of the simulation results using Linear Mixed Effect Modelling, progressively including variables in both fixed effects and random effects. We compared the Akaike Information Criterion (AIC) of each model to evaluate the fit of the model. AIC is an indicator of prediction error associated with a general linear model. It is an indicator of relative performance of a group of models; the model with the lowest AIC is concluded to be the one with least prediction error or best performance. With each iteration of the model, we selected the features most influencing the performance of the model and reducing the AIC of the predictions.

"

Line 253 (Equation 2): Is this a standard definition of Da? If so, please provide a citation. If not, please provide clarification as to how this equation represents the ratio of advective and reactive time scales.

We realize that this equation was not adequate to explain the derivation of Da in the scenarios. We will update the section with the following explanation on calculation of Da for the various scenarios. We also noticed there was an error in the equation which is rectified in the explanation below as well.

"

we used the Damköhler number (Da) to indicate the reaction regime for each reactive species. Da is defined as the ratio of the advective transport time scale and the reaction time scale as described in Eq. 2.

$$Da = \frac{\tau_{transport}}{\tau_{reaction}}, \qquad (2)$$

where, $\tau_{reaction}$ *is the characteristic reaction time scale and* $\tau_{transport}$ *is the characteristic transport time scale given by the breakthrough time of a conservative tracer in the domain. We adapted this definition* *and used Eq 3 below to calculate the apparent Da using values estimable in the field when* $\frac{C_{out}}{C_{in}} > 5\%$ *(adapted from Pittroff et al., 2017).*

$$Da = -ln\frac{C_{out}}{C_{in}}, \qquad (3)$$

with Cin as flux averaged concentration of a reactive species entering the domain, and Cout as flux averaged concentration of the reactive species leaving the domain. In case of $\frac{C_{out}}{C_{in}} \leq 5\%$, *we used Eq. 4 and Eq. 5 to derive the apparent Da of the chemical species*

$$\tau_{reaction} = \frac{-ln\,(0.37)}{-\,ln(\frac{Cy5}{Cin})} \times \tau_{y5}, \qquad (4)$$

$$\tau_{reaction} = \frac{\tau_{y5}}{ln(\frac{Cy5}{Cin})}, \qquad (5)$$

where, C_{y5} *is the concentration of the chemical species at the first cross-section (y = y5) when* $\frac{C}{C_{in}} \leq 5\%$, *and* τ_{y5} *is the breakthrough time for a conservative tracer at the same cross-section, i.e., y = y5.* $\tau_{transport}$ *in this case was the same as the breakthrough time of the conservative tracer in the domain (Eq. 6).*

$$Da = \frac{breakthrough\ time}{\frac{\tau_{y5}}{ln(\frac{Cy5}{Cin})}}, \qquad (6)$$

Thus, we were able to characterize reaction dominant system where Da > 1. We took the logarithm of Da to the base 10 ($log_{10}Da$*) to characterize the regime for each reactive species in each domain.*
For a scalable relationship addressing impact of spatial heterogeneity on reactive species removal, we conduct a simple linear regression analysis of species removal vs. residence time (both in relative units to the homogeneous reference cases) for different $log_{10}Da$ *ranges.*
“

Lines 265-270: These lines highlight a broader issue: What times were used for analysis and metrics of reactive processes? The flow field is clearly stated as being steady, but the concentration fields will be transient, and various times are mentioned in the manuscript. The breakthrough time is defined as the time at which Cout/Cin = 0.5 (for tracer), but what other times are considered for reactive species (they may not reach this value at the outlet)? This was confusing to me as a reader and should be clarified.

We agree that this aspect is confusing for the reader. We will rephrase the Data Analysis section to specify that (with the exception of the tracer tests) we are using the steady state concentration profiles for chemical and microbial species as well. Thus, both flow field and concentration fields are at steady state.
L237 onwards update to the text is as follows:

“(that is, DOC, DO, ammonium, and nitrate) from the domain in steady state conditions. Thus, while the chemical species entering the domain at the inlet were consumed at varying rates by the microbial species present in the system, the rate of consumption was constant in time in each domain in all flow regimes.”

Since we realized that spatial heterogeneity primarily resulted in reduced breakthrough times, we wanted to check if the changing breakthrough time is the lone driver for the reduction in removal of chemical species from the systems (proposed by Sanz Prat et al. (2015, 2016) as well), effectively reducing the problem to a zero dimensional (concentration changing in the time domain alone). So we used the analytical solution for first and zeroth order rate expressions to evaluate the reduction in removal with respect to reducing breakthrough time alone. However, we adapted the analytical solutions using the Damköhler number to generalize this discussion further (in Equation 5). We used a wide range of reducing breakthrough times (normalized by that in base cases, from 10% to 90% of breakthrough time in the base case) to solve Equations 5 and 6 (subsequently plotted in Fig. S8.

Technical Corrections:
Lines 58-60: awkward wording, suggest rephrasing

Thank you for the feedback. We will rephrase the sentence as follows:
"Microbial communities play a key role in these biogeochemical cycles since they mediate nearly all the naturally occurring processes that contribute to these cycles."

Lines 76-78: consider simplifying to "…representative of a system's chemical and biological species, and second…representative of a system's flow and transport pathways."

Thank you for the feedback. We will rephrase the sentence as follows:
"First, the reaction network should be representative of a systems' chemical and biological species, and second, the flow component of the model should be representative of a system's flow and transport pathways."

Lines 102-103: suggest grouping citations at the end of the sentence

Noted and addressed.

Line 153: e.g. seems out of place, consider deleting

Noted and addressed.

Line 155: aerobic should be all lowercase

Noted and addressed.

Line 162: necromass is one word, not two

Noted and addressed.

Line 163: complete the second half of the sentence by describing how microbes become immobilized (biofilms etc.)

Mathematically, the attachment of microbes depends on only the carrying capacity and concentration of immobile microbes (see sections A.3.3, L705 onwards). In a real system, we agree that immobilisation of microbes may be due to several reasons such as biofilms, interaction with the matrix etc. We rephrase the sentence as follows:

"Furthermore, the reaction network accounts for microbial attachment, in case of hospitable conditions, and detachment due to inhospitable conditions or velocity of the water (see section A.3.3). The detached mobile bacteria are transported by the flowing water."

Table 1: "longitudinal" is misspelled

Addressed.

Line 270: this is "the" same (add "the" to the sentence)

Addressed.

Line 280: switch scale and spatial in the sentence

Here, we intended to refer to the scale of the spatial scale sample. We will instead rephrase the sentence as follows for better readability:
"We explore flux-averaged concentrations of mobile species and spatially averaged concentrations of immobile species in 1-D, along the predominant flow direction, and explore the 2-D concentration heat

maps of the domain to compare the information lost when neglecting spatial heterogeneity at scales smaller than that of the sample."

Figure 2: Is a title necessary? The figure caption should cover the topic of the figure.

We agree, and we will remove the title in the revised manuscript.

Line 424: unbold sentence

Addressed.

Figure 4: Why is there a border around this figure?

This is a legacy error. We will remove the border in the revised figure.

Line 447: change to, "Since nutrient dynamics are..."

Addressed

Line 482: Is there a difference between dormant and inactive? The term "dormant" is used sparingly within the manuscript, so I'd suggest sticking to inactive and defining that this pool includes dormant microbes to avoid confusion.

We agree that we should be consistent with the terminology. We will use "inactive" consistently throughout the revised manuscript.

Lines 487-489: clunky sentence, suggest rewording

Rephrased in the revised manuscript:
"*The system may respond similarly to temporal fluctuations in groundwater velocities resulting from seasonal cycles as well."*

Figure 6: It's a bit unorthodox to present figures within the discussion section. Why didn't you introduce it within the results section and then reference it in the discussion?

We agree with the reviewer in that it is better to introduce Figure 6 in the Results section. We will rearrange the manuscript to accommodate this.

Line 618: Change to "The regression model links the..."

Addressed.

Line 631: Change to "… was considered when evaluating…" (delete "for")

Addressed

References

Desbarats, A. J., and Bachu, S.: Geostatistical analysis of aquifer heterogeneity from the core scale to the basin scale: A case study, Water Resources Research, 30, 673-684, 1994.

de Marsily, G., Delay, F., Gonçalvès, J., Renard, P., Teles, V., and Violette, S.: Dealing with spatial heterogeneity, Hydrogeology Journal, 13, 161-183, 10.1007/s10040-004-0432-3, 2005.

Pittroff, M., Frei, S., and Gilfedder, B. S.: Quantifying nitrate and oxygen reduction rates in the hyporheic zone using 222Rn to upscale biogeochemical turnover in rivers, Water Resources Research, 53, 563-579, https://doi.org/10.1002/2016WR018917, 2017.

Sanz-Prat, A., Lu, C., Finkel, M., and Cirpka, O. A.: On the validity of travel-time based nonlinear bioreactive transport models in steady-state flow, J Contam Hydrol, 175-176, 26-43, 10.1016/j.jconhyd.2015.02.003, 2015.

Sanz-Prat, A., Lu, C., Finkel, M., and Cirpka, O. A.: Using travel times to simulate multi-dimensional bioreactive transport in time-periodic flows, J Contam Hydrol, 187, 1-17, 10.1016/j.jconhyd.2016.01.005, 2016.

Turcke, M. A., and Kueper, B. H.: Geostatistical analysis of the Borden aquifer hydraulic conductivity field, Journal of Hydrology, 178, 223-240, 1996.

Welhan, J. A., and Reed, M. F.: Geostatistical analysis of regional hydraulic conductivity variations in the Snake River Plain aquifer, eastern Idaho, GSA Bulletin, 109, 855-868, 1997.