

General assessment: This manuscript presents a coupled numerical investigation of nitrate removal in heterogeneous hyporheic sediments. The modeling framework is compelling, and the findings offer valuable insights for improving the representation of biogeochemical processes in hyporheic zone models. However, before publication, the following points require clarification and revision.

Response to the general comments: We sincerely thank the reviewer for the constructive evaluation and for recognizing the value of the coupled numerical framework. The comments have helped us identify several places where the model assumptions and the physical meaning of our comparisons need to be explained more explicitly. We will revise the title and terminology, clarify the homogenization and boundary-condition assumptions, describe the single-biomass representation, distinguish direct biogeochemical effects from indirect hydraulic effects, expand the discussion of dimensionality and sediment stability, define the nitrate-removal metric, and correct the section numbering, Table 1, and notation. Our point-by-point responses are provided below.

1. Title Specificity: The title broadly refers to "nitrogen removal," whereas the study focuses specifically on nitrate removal via denitrification. Consider revising the title to "nitrate removal" or explicitly clarifying the scope of "nitrogen removal" in the introduction.

Response to comment #1: We agree and will revise the title to ‘Contrasting Roles of Microbial Heterogeneity and Sediment Heterogeneity in Controlling Hyporheic Nitrate Removal.’ We will also use ‘nitrate removal’ consistently when referring to the modeled denitrification sink and reserve ‘nitrogen cycling’ for broader contextual statements.

2. Homogenization Methodology: The equivalent homogeneous models utilize effective permeability tensors but employ arithmetic means for organic matter (OM) content. Please justify the use of arithmetic averaging for OM and clarify whether this approach conserves the total initial OM mass between the heterogeneous and homogeneous domains.

Response to comment #2: Thank you for this important comment. We will clarify the homogenization procedure in Sect. 2.2. Because OM content is represented as a mass fraction of the sediment, the equivalent homogeneous value was calculated as the volume-weighted arithmetic mean of the heterogeneous OM field. Given the uniform sediment bulk density used in the model, this averaging method conserves the total initial OM mass between the heterogeneous and homogeneous domains. In contrast, permeability controls the directional flow response and was therefore upscaled as an effective tensor following Durlofsky (1991). We will state this rationale and the OM mass-conservation property explicitly in the revised manuscript.

3. Boundary Condition Assumptions: The concentrations at the sediment-water interface remain constant throughout the 730-day simulation. Please confirm if the overlying stream is modeled as an infinite, well-mixed nutrient reservoir. A brief discussion regarding how this assumption might influence long-term nitrate removal estimates would strengthen the interpretation.

Response to comment #3: Yes. We will state explicitly that the prescribed sediment–water–interface concentrations represent an effectively infinite, well-mixed overlying-water reservoir

whose concentrations are not depleted by hyporheic uptake. In fact, the constant sediment–water–interface concentrations were prescribed to isolate the mechanisms controlling nitrate removal rather than to predict nitrate removal for a specific stream. The low- and high-nutrient scenarios represent persistently nutrient-poor and nutrient-rich conditions, respectively. Although the magnitude of nitrate removal differs between these scenarios, the identified mechanism remains consistent. This suggests that temporal variations in stream nutrient concentrations may affect the magnitude and timing of nitrate removal but would not alter the main mechanism identified within the tested concentration range. We will clarify this rationale and discuss the associated limitation in the revised manuscript.

4. Microbial Biomass Representation: Both aerobic respiration and denitrification are simulated using Monod kinetics with oxygen inhibition. Clarify whether these processes share a single functional biomass pool or distinct microbial guilds. If a single biomass variable is used, please acknowledge this simplification and discuss its potential impact on the spatial distribution of biogeochemical hotspots.

Response to comment #4: Thank you for this important comment. The present model uses a single functional heterotrophic biomass variable, X , for both aerobic respiration and denitrification. The dominant metabolic pathway is regulated by oxygen availability through the Monod kinetics and oxygen-inhibition term, representing facultative heterotrophic microorganisms under different redox conditions. This process-based simplification captures the first-order spatial separation between aerobic respiration and denitrification driven by oxygen and substrate availability. Distinguishing individual microbial guilds could further refine the predicted extent and intensity of the biogeochemical hotspots, but is not expected to alter the main redox-controlled pattern or the bioclogging mechanism investigated here. We will clarify this biomass representation and its scope in Sect. 2.1 and the Discussion.

5. Mechanistic Distinction: The interpretation heavily emphasizes progressive bioclogging near the sediment-water interface. Please more clearly differentiate between nitrate removal driven directly by enhanced microbial reactivity and that resulting indirectly from altered hyporheic exchange and extended solute residence times.

Response to comment #5: Thank you for this important comment. We agree that enhanced microbial reactivity and bioclogging-induced changes in hyporheic exchange represent two conceptually distinct pathways. In the present model, however, these pathways are dynamically coupled: microbial growth directly enhances nitrate consumption, while the associated biomass accumulation reduces permeability and modifies hyporheic exchange, solute delivery, and residence time. In fact, the purpose of this study is to evaluate the contrasting roles of microbial and sediment heterogeneity, rather than to quantify the individual contributions of microbial reactivity and bioclogging. Moreover, the current scenario design does not include a case with microbial growth but without permeability feedback, so these contributions cannot be separated quantitatively. We will clarify the two pathways in the Discussion while emphasizing that they jointly represent the spatiotemporally evolving microbial heterogeneity considered in this study. Their individual quantification will be identified as a topic for future investigation.

6. Dimensionality and Bedform Effects: The simulations are two-dimensional, representing repeating ripple structures. Discuss whether three-dimensional flow paths or non-repeating bedforms might introduce additional bypass routes around the clogged surface layer. This would clarify the applicability of the proposed mechanism to natural streambeds.

Response to comment #6: Thank you for this important comment. Three-dimensional flow paths and non-repeating bedforms may create localized bypass routes and modify the spatial pattern of hyporheic exchange. However, they may not fundamentally alter the mechanism identified here. Because microbial growth and biofilm accumulation are concentrated near the sediment–water interface, progressive bioclogging restricts solute transport and hyporheic exchange to shallow sediments, thereby reducing the influence of deeper sediment heterogeneity on nitrate removal. This feedback may also occur in three-dimensional and irregular streambeds, although the continuity of the clogged layer and the quantitative magnitude of its effects may differ. We will clarify this point in the revised Discussion.

7. Sediment Stability: The model assumes fixed sediment structure and streambed topography over 730 days. Please explicitly state that sediment mobility, bedform migration, and biofilm detachment are neglected. Given that these processes often limit persistent bioclogging in natural systems, adding a statement regarding this limitation is crucial.

Response to comment #7: We agree and will explicitly state that sediment mobility, bedform migration, and biofilm detachment are neglected in the model. Therefore, the simulated long-term bioclogging represents an upper-bound effect under a fixed streambed configuration. In natural streambeds, sediment reworking, bedform migration, and biofilm detachment may disrupt the clogged surface layer and reopen flow paths into deeper sediments. These processes would weaken the effect of persistent bioclogging and may consequently increase the influence of sediment heterogeneity on nitrate removal. We will add this limitation to Sect. 2.2 and the Discussion.

8. Section Numbering: Please correct the section numbering. "3 Results and Discussion" is followed immediately by "5 Conclusions," indicating that Section 4 is missing.

Response to comment #8: Thank you for identifying this error. '5 Conclusions' will be corrected to '4 Conclusions,' and all cross-references will be checked and updated.

9. Definition of Metrics: Define the total nitrate removal metric (RN) more explicitly. Specify the calculation method, integration period, and the physical meaning of the unit "g m⁻¹" within the context of a 2D model (i.e., per unit channel width).

Response to comment #9: We will add an explicit definition immediately before Table 3. The cumulative nitrate removal is calculated as $R_N = \int_0^T \int_{\Omega} -R_{NO_3} d\Omega dt$, with $T = 730$ d and Ω denoting the two-dimensional sediment domain. The volumetric reaction rate is integrated over the modeled cross-sectional area and a unit out-of-plane channel width; after unit conversion, R_N is reported in g m⁻¹, meaning grams of nitrate-N removed per meter of channel width over the

730-day period. We will also clarify the nitrogen-mass basis used in the calculation and apply it consistently in Table 3 and its caption.

10. Table 1 Corrections: Several entries in Table 1 require correction: (i) permeability units should be m^2 , not $m\ s^{-1}$; (ii) transverse dispersivity is mislabeled as α_L (longitudinal); (iii) clarify ambiguous range values and source labels; and (iv) remove the extraneous character in "1600c."

Response to comment #10: We agree and will correct Table 1. Permeability will be reported in m^2 ; transverse dispersivity is labeled α_T ; ranges are written with an en dash and separated by sediment structure (Brazos River: 10^{-12} – $10^{-12}\ m^2$; Massillon Sandstone: $10^{-11.5}$ – $10^{-10.3}\ m^2$); source labels are defined unambiguously in the table note; and '1600c' will be corrected to '1600'. We will also check dimensional consistency between permeability k and hydraulic conductivity K throughout the equations, text, and captions.

11. Notation Consistency: Ensure consistency in symbols and terminology across the text, tables, and figures. Address discrepancies such as OM vs. POC and X vs. X_h . All variables presented in Figs. 3–5 must be defined in the respective captions.

Response to comment #11: We agree and will complete a notation audit. 'OM' will be retained only as the general sediment-property term, whereas the modeled solid-phase carbon pool will be consistently denoted POC (C_{POC}). The single functional biomass will be denoted X throughout; X_h will be replaced. Permeability k and hydraulic conductivity K will be distinguished consistently. The captions of Figs. 3–5 will define every plotted variable and its unit: X (functional microbial biomass), k_{xx} (streamwise permeability component), DOC, DO, NO_3^- , R_{DOC} (DOC reaction/source–sink rate), and R_{NO_3} (nitrate consumption rate). The same symbols will be used in the governing equations, Table 1, and the main text.