Response Statement to Community's Comments (CC6)

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The authors thank the reviewer for the valuable comments. The manuscript has been revised by carefully considering all the comments. The changes are highlighted in the marked copy, and detailed responses to the reviewer's comments are provided below.

Comment #CC6:

I have some concerns about hybrid decisions. (1) You use element-based ADE transport, with a weighted molecular diffusion coefficient built from prescribed species fractions, while noting that fully coupled PhreeqcRM could update speciation and thereby Dm dynamically, but you don't do that in this study. (2) You also include a linear adsorption Kd term in the ADE, while stating that more detailed sorption could be handled by PHREEQC reaction terms. Please add a why this choice paragraph: is the goal robustness and speed? If so, say that explicitly and acknowledge the trade-off. Specify whether Kd is used in the case studies and, if yes, whether PHREEQC includes any overlapping sorption reactions.

Response:

Thank you for raising this important point. We agree that the modelling choices regarding the treatment of diffusion coefficients and sorption processes must be clearly stated.

In this study, the species fractions ω_i used to compute the effective molecular diffusion coefficient $D_m = \sum_i \omega_i D_m^{(i)}$ are prescribed as constants. Although the numerical architecture can, in principle, be extended to dynamically update ω_i using cell-wise speciation results from a geochemical module, such functionality is *not activated in this work*. Dynamic coupling would require code-level modifications and recompilation whenever the reaction network is altered, which substantially reduces model reproducibility. Moreover, as D_m is formulated as a weighted average of species-specific diffusion coefficients, its variation under typical speciation changes is expected to be limited and is unlikely to affect the principal chemo-mechanical trends examined here. We have revised the manuscript to explicitly state this rationale.

Regarding sorption, although the governing equation is presented in a general form that can accommodate a linear distribution coefficient K_d, *all simulations reported in this paper set* $K_d = 0$; that is, no equilibrium sorption or retardation is considered. Furthermore, no PHREEQC-based sorption reactions are activated in the present study, and therefore there is no overlap or double-counting between an ADE retardation term and geochemical reaction processes. The statement that PHREEQC can represent more complex sorption mechanisms has been clarified as a future extension and is not part of the current implementation.

[Deleted content:] ~~The reaction module (PhreeqcRM) can also handle more complex sorption processes, including nonlinear (Freundlich or Langmuir) and competitive or kinetic adsorption–desorption, if specified in the chemical database.~~

[Added new content:] *The PHREEQC reaction module is, in principle, capable of representing more complex sorption mechanisms (e.g. nonlinear Freundlich or Langmuir isotherms and competitive or kinetic adsorption–desorption), but these functionalities are reserved for future extensions and are not used in this work.*

[Line 208–209]

[Deleted content:] ~~Within the coupled hydro–mechanical–chemical framework, the relative fractions of aqueous species ω_i associated with each element can be dynamically obtained or prescribed as constants. When executed in a fully coupled mode with PhreeqcRM, the speciation module automatically determines species concentrations in each cell and time step, allowing $\omega_i = Y_i / Y_{\text{element}}$ to evolve with local chemistry. This allows the effective diffusion coefficient $D_m = \sum_i \omega_i D_m^{(i)}$ to vary consistently with reaction, pH, and ionic strength. The species-specific diffusion coefficients $D_m^{(i)}$ are retrieved directly from the Phreeqc database via the `-diffusion_coefficient` entries, ensuring a thermodynamically consistent parameterisation for both static and dynamic simulations.~~

[Added new content:] *Within the coupled hydro–mechanical–chemical framework adopted in this study, the relative fractions of aqueous species* ω_i *associated with each element are prescribed as constant parameters, and the effective molecular diffusion coefficient is evaluated as* $D_m = \sum_i \omega_i D_m^{(i)}$*, where the species-specific diffusion coefficients* $D_m^{(i)}$ *are obtained from the PHREEQC database through the* `-diffusion_coefficient` *entries. Although the present numerical architecture can, in principle, be extended to dynamically update* ω_i *using cell-wise speciation results from a geochemical module, such functionality is* not activated in this work*, because dynamic coupling would require code-level modifications and recompilation whenever the reaction network is altered, substantially reducing model reproducibility; moreover, as* D_m *is formulated as a weighted average of species diffusion coefficients, its variation under typical speciation changes is expected to be limited and is unlikely to affect the principal chemo-mechanical trends investigated here.*

[Line 208–209]