MF6PQC documents an important linkage between two widely used U.S. Geological Survey codes, MODFLOW 6 and PHREEQRM. A previous incarnation of this particular linkage was "MODFLOW/MT3DMS-Based Reactive Multicomponent Transport Modeling" (https://doi.org/10.1111/j.1745-6584.2003.tb02588.x) in 2003. A more comprehensive listing of its history is available from https://www.pht3d.org/pht3d_about.html. Since MT3DMS is no longer actively supported and the core version of MODFLOW is now MODFLOW 6, the work presented in "MF6PQC v1.0: …" represents an important software update since it makes use of the latest MODFLOW (i.e., MODFLOW 6) and PHREEQC (i.e., PHREEQRM) codes.

The manuscript does a nice job of comparing output from MF6PQC with other established solutions, that is, previously published results from a PHT3D simulation (the first example problem), the Hamann et al. (2015) study (the second example problem), and MIN3P (the 3rd and 4th demonstration problems). Having established the veracity of MF6PQC's results by comparing simulated outputs to established solutions, the authors then use the code to evaluate the impact of accounting for MODFLOW 6-PHREEQRM feedbacks that account for (1) density only, (2) hydraulic conductivity and porosity only, and (3) density, hydraulic conductivity, and porosity. The authors do a nice job showing how the feedbacks in shared variables, for example density, has important implications on the model output when the models are tightly coupled (i.e., linked by a Picard iteration that uses a tolerance on shared values to determine when the integrated code has converged and can therefore move onto the next transport step). Furthermore, solutions are archived on Zenodo for reproducibility, and not just of the worked examples, but for other to see how the codes were linked for application to areas that might be in reader's jurisdictions.

This manuscript merits publication. It provides an updated linkage between two important USGS codes that have recently been reconfigured to align with Basic Model Interface principals. Moreover, linking these two codes results in a synergistic coupling that can help address complex hydrogeochemical questions in natural settings with important water quality implications. As with most submitted manuscripts, there are some issues that should be addressed and are listed below.

Specific Comments:

Lines 105 to 110: Since porosity is updated, this implies updates to storage, specifically to the specific yield term. When changes to storage occur, has thought been given to how this impacts the accounting of changes in mass in the compressible storage component, "Ss"?

Line 274: After having read the discussion on lines 266-274 and having looked at figure 8, the abrupt change in porosity and hydraulic conductivity at about 0.5 m that doesn't

change from the 100 yr to 300 yr is hard to understand. Please provide a better physical interpretation of why the abrupt change in hydraulic conductivity remains relatively unchanged from 100 to 300 years when prior to that, there seems to be a more smooth evolution of the aquifer properties (the 10 yr line).

Technical Corrections:

Line 50: Consider including the Hughes et al. (2017) MODFLOW Framework citation next to the two Langevin citations (https://doi.org/10.3133/tm6A57)

Line 54: Next to the Bakker et al. (2016) citation, you might also consider including the more recent FloPy citation (https://doi.org/10.1111/gwat.13327)

Line 88: where is says "…per unit solution volume, …" it would be helpful if perhaps you parenthetically included a mathematical expression of what you are talking about, perhaps: "…per unit solution volume (θV_{cell}), …"

Line 98: Strikethrough text suggest a deletion, bold font-type test recommends an insertion: "Porosity is updated ~~from~~ **based on** the net change in…"

Line 98: Please clarify what you mean by "inert-framework volumes"? Is this meant to suggest the aquifer material is non-reactive?

Line 123: Should "A python driver…" instead be "A python script…" It seems more clear to me, but maybe that's just me.

Line 123: You mention that FloPy was used to generate the MODFLOW 6 input; is there a similar library for preparing the PHREEQRM files that are specific to a particular application, or do users of MF6PQC need to be prepared to generate a few PHREEQRM input files by hand?

Line 124: Please be clear that the python script is advancing both MF6 and PHREEQRM.

Line 124-125: The sentence that reads "The flow, transport, and reaction modules retain their native solvers" could be more clear by stating which processes are solved by which codes. For example, "Flow, advection, and dispersion are solved within the MODFLOW 6 hydrologic simulator whereas chemical reactions are solved by PHREEQRM."

Line 123/125: On line 123, the authors use the term "Python driver" and on line 125 the term "coupling layer" and then I noticed on line 133 you use simple "The driver." I'm thinking these terms are each referring to the same python script that is used to establish

the linkage between the two models. Recommend using consistent terminology to reduce the potential for confusion.

Line 129: What is meant by "boundary-solution concentrations"? Please clarify. Is "solutions" referring to the result solved for (i.e., a numerical solution) or is it referring to the fluid? "Solutions" is used a second time on this same line and I get the sense it is referring to a PHREEQRM term that needs to be defined for the reader.

Also on line 129: the term "…FloPy models" doesn't make sense. FloPy is a python library for creating, running, and post-processing a MODFLOW 6 model, but it is not a model itself. Do you mean to refer to the "…MODFLOW 6 GWT models"? If so, you might also want to cite the GWT documentation (https://doi.org/10.3133/tm6A61).

Line 130: "…reactants can be assigned by spatial zone." What does this mean in practical terms? Do modelers need to be prepared to assign every MODFLOW cell a "zone" that defines how chemical reactions will be handled in PHREEQRM? Where is the spatial zone defined, in native PHREEQRM input or somewhere else?

Line 130: "…component concentrations for the individual GWT models." You might consider highlighting that each component, or "species" (if we were to stick with the terminology established long ago for MT3DMS: Mass Transport in 3-Dimensions for Multiple **Species**"), requires it's own GWT model. That is, each GWT model is capable of simulating only one species and so a simulation with multiple species requires multiple GWT models. On it's own, MODFLOW 6 cannot currently simulation reactions among the different GWT models.

Line 135: "…the same physical interval."? Do you mean the same "temporal" interval? It's not clear what the authors mean.

Figure 2: Figure might benefit from a legend that defines the different symbols. For example, ΔV_{min} has not been defined. Also, in the yellow-shaded box, what is "D", is this molecular diffusion? If so, elsewhere you've defined D_0, D_p, D_e, and D_i, but I don't recall seeing D. Maybe it's not related to molecular diffusion? Finally, what does "commit" mean? Perhaps you mean to say something like "advance to the next transport step"?

Line 147: "The rightmost operator is applied first…" It is not clear what this is referring to? U^n in equation 6? Something else. Please add a variable or equation reference to help clarify what you are referring to.

Line 148: "…written back…" implies that variables I thought were shared in memory are possibly written to a text file that is read by the other half of the integrated code. Perhaps

the authors mean "passed back to MODFLOW 6 via pointers to shared memory"? I'm asking that the authors please be as specific as possible for clarity.

Line 150: With regard to "Strang splitting," would the authors please add some text that would clarify how it works in a practical way. In other words, if I were to use Strang splitting in my application, would the reacted concentrations that are written to the output at the end of the transport step be representative of the time at the halfway point of the transport step?

Line 154: The authors might consider adding a reference to the end of the following sentence "…with the ordering of transport and reaction calculations **as shown in Author et al. (20xx)**." Would it be Lu et al. (2022b)?

Line 156: "…can also…" should be "…also can…"

Line 157: recommend changing "…restores the…" to "…starts with the…"

Line 157: There is a seemingly abrupt change at the end of the line "Let s^j be …" s^j has not been introduced yet, so why is it entering the discussion without being defined first. You might need to introduce the equation followed by the typical way of defining the terms, for example "where x is such and such, y is this, s^j is the concentration source term at iteration j, etc." I think this is a more clear way to introduce the terms.

Line 160: Should equation 8 appear on two lines, the part after the comma is just rearranging terms, so maybe provide the first equation (left of the comma) and then state "after rearranging terms to solve for s^j we get…"

Equation 8: What is C_B^j? I didn't see where this was defined.

Lines 163/164: This seems like the first "Verification Test Problem"? Should the section header that appears on line 178 be moved here instead?

Line 168: add figure reference to "…with the analytical solution at 0.5 yr **(Figure 3A)**."

Line 169: add figure reference to "…and 0.00338 for SNIA **(Figure 3B)**, …"

Line 170: add figure reference to "…and 2455 (**Figure 3C)**."

Line 179: What about enumerating the examples with integers (bold text represents recommended insertions: "We evaluated component transport and mineral reactions against **(1)** PHT3D, **(2)** them examined density-driven circulation and …" and "Comparisons with **(3)** MIN3P…" and finally "…before **(4)** the combined feedback application in …"

Line 183: Perhaps the section header should include something like: "**Example 1:** Reactive transport with fixed physical properties". In addition, I'm wondering if it is actually example 2 since a comparison was documented on line 165?

Line 188: Perhaps also add the Courant number.

Line 201: I recommend adding a subheader to help make it clear a new example is being described. Perhaps "**Example 2: 2-Dimensional heterogeneous flow with reactions**".

Line 202: Suggest inserting the text shown in bold, "The 200 m x 50 m domain contained 80 x 40 cells **as shown in Figure 5A**."

Line 205: Regarding the use of an example that employes the TVD scheme: I think it's important to highlight that there are some significant differences between the 3rd order TVD scheme that's in MT3DMS (and therefore PHT3D) and the 2nd order TVD scheme that's used in MODFLOW 6's GWT model.

Line 209: Suggest changing, "…the head RMSE between…" to something a little more clear, such as "At 500 d, the RMSE of the hydraulic head residuals between …"

Figure 5: In 5A, need a label for the x-axis: "x (m)". Also, it is hard to see the source area contour where it overlaps with the dark shaded area. Suggest moving the colorbar legend that appears underneath 5A to the right hand side of the image. In 5A, I can't see any difference between the MF6PQC contour? and the PHT3D contours? Perhaps they are directly overlapping one another, which would be great, but I can't tell that. For figure 5B through 5M, there's a similar problem of not being able to distinguish between the solid and black-dashed lines. I suspect they are well-aligned, but I can't tell (in a printed version of the image), everything looks dashed.

Line 217: Consider changing header to something like, "**Example 3. Density…"**

Line 219: Considering adding a figure reference at the end of the sentence, "…was 100 m long and 10 m deep **(Figure 6A)**"

Line 224: The discussion about refinement is unclear. Is it referring to the horizontal or vertical refinement. Please be more specific to make this part of the discussion very clear.

Line 227: "…solutes removed by the conventional discharge calculation…" I'm not clear on what's happening here. I thought an MT3DMS user could set the concentration of the ET simulated by the MODFLOW ET package to zero such that there would be no solute removed with the removed water (that's my understanding as a former MT3DMS user, but I will go back and review)? In MF6, a user can use the AUXMIXED option in the SSM package (of a GWT model) to set the concentration of water removed by ET to zero (i.e, remove pure

water). For example, from the MODFLOW 6 I/O guide: "the AUXMIXED option is designed to work with the Evapotranspiration (EVT) and Recharge (RCH) Packages where water may be withdrawn at a concentration that is less than the cell concentration." So are you using something else to remove water, having to calculate how much solute mass was removed, and then put that back? Seems overly complicated.

Line 228: Suggest the following edits where bold text indicated a recommended insertion: "…0.1 yr time step and **accounted for** density feedbacks ~~alone~~."

Line 235: "Mineral precipitation became…" Recommend adding a figure reference earlier. Also, might it be possible to show a spatial plot that differences the before and after concentration of precipitated mineral, for example calcite, to further back this sentence up? Or did you already try such a figure and it didn't work well? Also be sure to add the figure reference to the end of the sentence if you do add a new spatial plot.

Line 236: suggest a few minor edits (where bold indicates insertion): "…gymsum occurred in **a** more concentrated region~~s~~**, specifically the righthand side of the domain**, and halite…"

Line 239: For the sentence that starts, "This pattern forms a two-dimensional…", I'm unable to follow the reasoning based on the figures available. You might consider adding a 2D figure that zooms in on the upper 0.5 m (since that is the zone you are commenting on) to help with this part of the writeup because I'm unable to follow what's being described. I don't see an evaporite "bull's eye".

Line 249: Might rename the section header: "**Example 4: Mineral reactions…**"

Line 259: "Both cases used implicit transport-reaction coupling…" Is this implying SIA? If so, recommend explicitly stating that.

Line 299: Suggest updating the section header to something like: "**Example 5: Application…**"

Line 305: Suggest adding a figure reference, for example (Figure 10A), at the end of the sentence that starts on line 305.

Line 308: Is the value "110 m" correct? If this is a drain elevation, it seems it would be above the top of the model. Does the recharge build up considerable head within a confined simulation? If so, you should better explain that.

Line 311: The reference to Figure 10C precedes a reference to Figure 10B. You might reorganize the figure so that its subfigures are referenced in order within the text.

Line 325: You might renumber the section header to instead be a subheading under 4.1. So for example, you might choose to convert the section header 4.2 to 4.1.1 and section 4.3 to 4.1.2

Line 325 and following: The authors use "K" to represent two distinct quantities throughout this section, which is confusing. In many places, "K" is shorthand for hydraulic conductivity and in other spots it references the periodic table's symbol for potassium. For example, see Figure 11. In subfigure "e", K refers to hydraulic conductivity but in subfigures b, d, f, & h it would seem to be referring to dissolved potassium concentrations.

Figure 12: Very nice figure!

Line 372: Suggest augmenting "medium" with "…**aquifer** medium…"

Line 381: Please clarify what this sentence is comparing? Is it comparing S_{11} to S_{01}? And is the next sentence referring to S_{10}?

Line 386: "Existing FloPy ~~models~~ **scripts** can…"

Line 391/392: The sentence "The comparison with MF6RTM…" Which example problem compares MF6PQC with MF6RTM?

Line 405: You might want to provide a reference for the GWE model: https://doi.org/10.1111/gwat.13470