13: hydrolgy processes

32: spatially implicit: what this means is not clear.

- abstract: not clear weather the verification with Crunch is part of the paper

42: microbes ?42: more general than CO2: gases fluxes?

59-60: unclear sentence

38-64: there are other references which could be worth citing

64: remove "that" 65-69: this is a pretty long sentence.

70: MacQuarrie and Mayer not sure this is the best citation in this sentence.

128: solar radiation?

- Figure 1 : beautiful figure!

- 144: kinetically controlled or at equilibrium

- 145: no eg for kinetically?

- Figure2 : resolution is not great. it may be due to the journal processing of the figures to generate this file. please be careful with resolution.

- eq(1) and (2) : why is the porosity there? in the previous figure caption, it says "h" refers to "head", so i expect meters. in typical Richards equation, this would yield a specific storage coefficient, for example. but then in the paragraph, h is the water storage. What does that mean? what units? L of water/ L of porous medium?

- need to be consistent between figure 2 caption and this equation.

- please say where this reaction comes from and define the different terms clearly and properly with units.

- Looking at Li 2019 RiMG, the storage is defined in m of water, but the porosity does not appear.

- eq(2) : sum ij to 1? this isn't clear. why is j <= 3? this notation is unclear. i'd rather have a sum over symbolic "neighbors" than not defining i, j properly and have a "sum up to 1"

- in every equation, "shallow", "lateral", and actual english words should not be in italic. \textrm{} or \mathrm{} in LaTeX, or straight formatting in word.

- 190: this should not be a new paragraph.

- Eq (3) and (4): this sugests that q has units of m3/s. In equations (1) and (2), if h is in the units of meters, q should have units of m/s.

Anyway, there are inconsistencies between the equations. This could be dealt with simply by removing "A" from the equation (and in the text).

- 191: K\_infil ^shallow : this is an infiltration? from the form of equation (3), it looks like a hydrualic conductivity.

- 192: in the vertical direction? this suggests some kind of anisotropy: is this the case?

- all the notations with sub and superscripts are really not clear. this should be significantly improved for readability. maybe call q\_rechg with a new symbol R, not write "shallow" on every single term, ...

- 198: K\_eff H\_ij ^shallow --> very heavhy notation again. why the need of H\_ij. I guess it could simply be written as K\_ij (every other K has ^shallow), i don't see why a distinction is required for the "effective".

- 199: now the subscript H is used to denote horizontal. I would suggest simply K\_ij to say averaging between two adjacent cells. and if there is some anisotropy, maybe just distinguish using subscripts z, or x, to avoid confusion between all these symbols.

- eq(7): similar problem. subscript "i" is not defined.

these equations all adopt some form of discretization and are written for a volume "i". maybe some of the notation and clarity issues would be diminished if they were not written in a discretized fashion, hence removing all the "i" and "j" from the equations.

Because in theory, a subscript "i" should be added to every other variable to be consistent.

- also if there are indeed written in a discretized fashion, somehow volumes and surfaces should appear. they don't. or they're hidden within the variables. but it's hard to say as almost no units are given and variables are not clearly established.

- eq(8): same as equation (5). Typical Darcy's law, it could be given once, and text could specify where it is used (if needed).

- eq(9) --> please explain the denominator

- line 235: why area fraction instead of volume fraction?

- 238: infiltration hydraulic conductivity --> please change this denomination. hydraulic conductivity of a certain porous media, but not infiltration.

--> section 3.1 shoud be significantly improved. this section does not introduce many new concepts, but somehow is surprisingly unclear, due to very heavy notations, repetitions, ...

Section 3.2 here in equation (14), the discretization factors (Vi and Aij) do appear. Why not in the previous ones?

- (14) the indices around the sum symbol are pretty heavy. maybe just put sum over j(neighbours), and explain in the text than j refers to the neighbours of cell i. but N\_i,1 to N\_i,x is confusing
- (14)/256: all units of distance within gradients have been expressed with "d" or "D" and here it's I\_ij. maybe some consistency could be appreciated.

- 250: + gas volume, i guess (or simply remove the parenthesis, i don't think it's important).

- 250: put the m3 in [] somewhere else, like where Vi [m3] is the total volume of gridcell i.

- 252: index of elemets sharing surfaces --> it's unclear and i don't think it adds anything

- 255: "combined". for consistency i would suggest to stick with "mean" as was done for hydraulic conductivities (is it harmonic?)

- 257: agree that here, q has m3/s, because Aij has been incorporated. Be careful about consistency with previous section and equations where it seemed to me that q was m/s (but again, it was not very clear).

- 261: microbes

- 265: precipitate as carbonate materials?

- 265: transition between sentences can be improved. "it can oxidize into co2. or it can precipitate. hence it can release co2". it would make more sense, i think, if it was grouped differently: " oxidize into CO2 which can be released back to the atmosphere or surface water. or it can precipitate".

- 266: chnage CO2 level (not changes, i think).

- Figure3 : resolution is not great again. But nice illustrating figure, it is helpful.
- 276: maybe give chemical form of ammonia. why is No2- within parentheses?

- It is weird that these paragraphs are part of a section called "equations". Maybe another section after line 258 "Biogechoemical processes"?

a first "paragraph" about description, then the "paragraph" describing kinetics (294) (And maybe section 3 should be modified to governing equations and processes?)

- eq(15)/ line 303 : why the subscript "c5h7o2N" and not simply microorganisms?

- can you give an example of D and A (electron donor and acceptor)? are they linked to the three pools, A and D being the intermediate stages?

- line 307: espectively; "they are the concentrations at which half of the maximum rates are reached for the electron donor and acceptor respectively" i think this can be omitted. it's part of the reference and is fairly well understood or self-explanatory.

- equation (16) could be summed up in one equation with one symbolic term like (Product)\_inhibiteurs K\_inh / (K\_inh + C\_inh) and then in the text examples could be given.

- 334-336: with little pore space for air. i'm not a big fan of this terminology. and the overall paragraph is not very clear. I would simplify like

important under conditions where electron donors and acceptors are limited, e.g. anoxic conditions for O2. Under conditions org carb and o2 are abundant, SOM rate loaw ... eq(19) and the following.

- eq(19) and 340. need to change notation for  $\mu$ \_max. it's not the same unit, it does not bear the same meaning.

- eq(19): which surface area? m2 of what?

- 348: if it is often used, i guess you could include some citations here :)

- 351: accounted for

- eq 20: i would suggest to merge equations 19 and 20.

- eq 20: bm is the declining coefficient? It's simply a characteristic depth. "declining coefficient" is a weird denomination.

361: CVODE? what do the CV stand for?

- you do not specify how you solve set of equations 14 (reactive transport) which are arguably the most complicated to solve.

- 374: at a range of reaction complexity levels? "a variety of transport conditions" is a bit overselling what has been done in the SI.

In my understanding, there is only one transport condition and 3 investigated reaction networks.

- there are 2 secions "4"?

- 398: i'd rather say something like "negligible, as the associated evolution of hydrological parameters".

- fig 4: you mention a lot of things which were not discussed in the model section. for example, thermal effects (evapotranspiration, solar radation, air temperature) --> how do they impact what has been discussed.

Basically, most of the complexity from figure 1 and figure 4 was absolutely not addressed in the model section. are there some missing references?

- temperature effects: how do they impact thermodyanmic constants, evaporation, ...

- ET: how do you compute that?

- Nash Sutcliffe efficiency? can you explain what that is? or refer to some work regarding that?

- 560: issue within the chemical species

- eq 25-26: some references are needed

- structure of the paper needs to be revised. a huge amount of new variables and informations are given within the applications.

- results: generally nice figures and results. but the leadup to here does not give the results the credit they deserve.

## ----- SUPPLEMENTARY

Table S2 : given rates are surface rates, but no surface area are discussed and the kinetic law is missing.

- it would be nice to add the Saturation index of the initial solution and boundary condition solution with respect to apatite.

- figure S2c: looking at h+ concentration on (a) after 1 residence time, it looks like it at 1e-8 mol/L (and going down). in figure s2c, downstream h+ concentration looks higher. as its probably in equilibrium with apatite, it's surprising that this value is higher than in figure S2(a). can you comment?

- table S3:

-units of rate constant.

- what is X\_mio?

-what is CH2O(s) ? s for solid? how do you define their concentrations? what does it represent? i would understand if it represented a surface area but this seems odd.

S12 --> what about flow/transport setup in this simuation? same as for the first? should be indicated

line 128: i think the reference is Figure S6. - same transport than previously?

table S5: same comments than for table S3

S2. what's the exponential factor?

Figure S9: (a) it's really not clear what is observed .. precipitation is the grey on the top, I guess. could you write "precipitation" in gray then? Please include evapotransportation (ET) in the caption.

the word "precipitation" in the middle of the frame is surprising.

- Figure S9(c): there is more water in the unsaturated than in the saturated part?