Interactive comment on “Caffeine vs Carbamazepine as indicators for wastewater pollution in a karst aquifer” by Noam Zach Dvory et al.

Noam Zach Dvory et al.
nzd@etgar-eng.com

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Answers to Reviewer #2 comments:

Thank you for the feedback. Your recommendations were helpful and insightful. All of the comments have been addressed, and the paper was edited accordingly.

General comments:

Figures 1, 2 and 3b are taken from the previous manuscript with no or very little modification. In addition to the overlap as indicated by the editor this should be addressed (Figure 1 is probably the most critical one and should be slightly modified, also to avoid any copyright issues).

Answer: As suggested by the reviewer, the Fig. 1 was modified. In the caption under figure 2 (Figure 3 in the revised manuscript) we indicate that the figure references Dvory et al., 2018a. Figure 3 (Figure 4 in the revised manuscript) includes CAF concentration unlike the previous paper published in WR.

Content-related comments:

1. (p6): For the sake of completeness please add units to the description of the equations.

Answer: The units were added.

2. (p5): The section "Numerical model" would greatly benefit from a conceptual sketch of the model framework (the Water Research paper provides a conceptual model of the hydrogeological system only). The model is quite complex and as the authors have already limited the model description here. A conceptual sketch would also allow readers to understand more aspects of the model without having to read another paper. This would also help to make the paper stand out slightly more compared to the WR paper.

Answer: As suggested by the reviewer, we added an additional figure (fig. 2) that we hope that will assist the readers to understand the model framework.

3. (p5, line 28): From the description here it is not entirely clear to me if both the 1D and 3D part of the domain are subject to a multi-continuum coupling. In this sense also the terms high and low permeable region are (from a conceptual point of view) associated with different compartments of the aquifer. High permeable regions in the vadose zone are possibly (enlarged) fractures and to a limited degree former conduit systems depending on the long term evolution of the system, while in the 3D part the high permeable regions are commonly the conduits. This should be clarified, possibly also in conjunction with my previous comment to add a conceptual sketch.
Answer: In the revised manuscript we clarify that multi-continuum coupling was done for both the unsaturated and saturated zones, which is also shown in the conceptual sketch (new Figure 2). In the mathematical model, fractures and conduits belongs to one continuum, while porous matrix to another.

4. (p6): In addition to the conceptual sketch I think a figure showing the discretized model domain including boundary conditions (both for the large and small model) would be adequate to be added to the section "Mathematical model setup".

Answer: We present the boundary conditions in Figure 2 (according to the revised version), however we prefer not to show the finite-differences discretization of the domains because it's technical and overloads the figure. The sizes of numerical grids are mentioned in the text.

5. (p7, line 15-19): To what extent does the vadose zone possibly affect the (bulk) dispersivity? The chosen approach is common for saturated systems but may be affected by the vadose zone which imposes an additional transformation/dispersion of the signal. I understand that this is a very difficult topic and would only ask for a brief comment if this might be the case (or not if the authors can clearly rule this out). In this context the authors mention that CBZ is stored in the vadose zone (on page 14, line 11), hence I would expect an influence.

Answer: We agree with the reviewer. The dispersivity parameters could be different for the vadose zone and groundwater, however, with the quality of data we have (breakthrough curves in one observation well) we can only obtain a lumped parameter for both unsaturated and saturated zones. We added the following note in section 3.2 (page 10, lines 11-16): "...These values, calculated in this study, represent combined vadose zone-groundwater model characteristics. Even though the presence of air phase can influence the physico-chemical processes of contaminant transport and transformation given the quality of dataset available (breakthrough curves in one observation well) we can only obtain lumped parameters for both the unsaturated and saturated zones. The effect of variable water saturation on pollution dispersion and degradation is accounted for by multiplying these parameters by the water content (equations (1) and (2)). " CBZ is stored in the vadose zone mostly in low permeability sites (matrix) and the rate exchange between matrix and conduits influences the transport. We added a clarification on this in section 3.4 (page 15, lines 13-15): "The tail of the low CBZ concentration during the dry season is a result of low saturation in the vadose zone. This reduces the hydraulic conductivity and the exchange between matrix and conduits, resulting in low CBZ transport rates toward the aquifer.”

6. (p12): Are the parameters $\lambda$ and $K_d$ defined for both the 3D section and the 1D vadose zone? This information should be added. Please also see my comment 3. Is the degradation of caffeine affected by the presence of an airphase? If this is the case then this should be briefly discussed either here or in the discussion section.

Answer: $\lambda$ and $K_d$ were defined for both vadose zone and the aquifer. We added a remark to this effect in section 2.4.3 (page 7, line 12) and briefly discussed this in section 3.2 (page 10, lines 11-15)

7. (p13, Fig4): I am a bit confused by both sensitivity analyses but may have missed some information in the manuscript. I would expect an opposite behavior for $\lambda$ as well as $K_d$. I would expect lower peaks (and low tailing, i.e. generally a decline in mass) for higher values of degradation. The same applies for the distribution coefficient (which to my knowledge is commonly defined as activity of solid/aqueous phase). Here I would expect lower peaks for higher values of $K_d$ as CAF tends to be in an sorbed state. In Figure 4a it is difficult to see where the peak of parameter combination 6 is (only the tailing is clearly visible). A different color (gray or colored) for the attested values (both in A/B) could help to enhance visual clarity.

Answer: Thank you for this important remark. The legend in this figure was wrong. We corrected the legend and the text accordingly. We also changed this figure (Figure 5 in the revised manuscript) to make it more coherent by distinguishing between the
different graphs lines.

8. (p14, line 5): Is this correct? I would expect low background concentrations to be beneï¬¬cial for the detection of a new signal.

Answer: Yes, it is correct. When the new event has low concentration levels that can occur as a result of surface or subsurface dilution it is more difficult to detect it from previous background concentrations.

Typographic corrections:

I am not a native speaker and can only partially comment on proper grammar. The following are mostly typographic corrections and recommendations to enhance the comprehensibility.

1. (p3, line 6): Maybe rephrase. Do the authors mean that Sorek creek watersheds accounts for 88km2 within the study area or that the Sorek creek study area is 88km2 in size?

Answer: The correction has been made as follows: “The Sorek creek watershed drains approximately 88 km2 in the study area and is located west of the city of Jerusalem, Israel (Fig. 1)”

2. (p3, line 19, 20): This may be journal-specific but commonly only numbers exceeding 12 are spelled out.

Answer: The correction has been made.

3. (p6-7, line 28/1): I assume the authors mean main memory not the CPU cache. Possibly rephrase as “owing to a lack of main memory”.

Answer: This part of the sentence was removed from the paper.

4. (p7, line 4): Is the grid becoming finer towards the top or the bottom of the domain? Possibly rephrase to clarify.

Answer: In the revised manuscript we indicate that the grid become finer towards the top of the matrix (the ground surface).

5. (p12, line 11): “…assigning λ = 0 in the matrix,...”

Answer: The sentence is correct. Thus, we tested a scenario in which CAF degradation is neglected in the matrix.

6. (p13, Fig. 4): Please match the font/fontsize of the insets in A and B (lambda and Kd values)

Answer: The correction has been made.

7. (p14, line 23): “A quasi 3D dual permeability...”

Answer: The correction has been made.

8. (p14, line 25): I think it should be “calibration with monitoring data...”

Answer: The correction has been made.

Please also note the supplement to this comment: https://www.hydrol-earth-syst-sci-discuss.net/hess-2018-426/hess-2018-426-AC2-supplement.pdf

Fig. 1. The upper Sorek Basin monitoring sites and flow and transport simulation domains (after Dvory et al., 2018a; aquifer boundaries from Dafny, 2009)

Fig. 2. Model conceptual sketch
Fig. 3. Time series data observation and calculation (after Dvory et al., 2018a). (A) Tzuba Meteorological station daily precipitation rate; (B) Dam runoff flow; (C) Sewage surface flow; (D) Measured and simu

Fig. 4. (A) Observed and simulated BTCs of CBZ and CAF in EK11; (B) Relative concentration variations of CBZ and CAF in EK11 (CBZ data from Dvory et al., 2018a).
Fig. 5. Simulated CAF sensitivity to parameters changes (A) the degradation rate and (B) the distribution coefficient. The insets show the effect of parameters on RMSE.