Hydrol. Earth Syst. Sci. Discuss.,
doi:10.5194/hess-2016-663-AC2, 2017

Hydrology and Earth System Sciences
Discussions
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Interactive comment on "Transport and degradation of perchlorate in deep vadose zone: implications from direct observations during bioremediation treatment" *by* Ofer Dahan et al.

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Received and published: 19 March 2017

Reply to reviewer # 2 comments on the manuscript:

Transport and degradation of perchlorate in deep vadose zone: implications from direct observations during bioremediation treatment

We would like to express our great appreciation to the reviewer comments and believe that we can address all questions and comments raised in this review. General comments

Comment: The major concerns are: i) the absence of any quantitative modelling of the water transport and/or the perchlorate pollution plume during the infiltration experiment; ii) the absence of any uncertainty assessment. Hypothesis related to the fate of the perchlorate plume are indeed subjected to the hypothesis of mass conservation and representativity of the singular sampling. These strong hypotheses can only be considered acceptable in the present case if the experimental results are compared with some quantitative modelling that are built on mass conservation principles (using e.g. a numerical water and solute transport, or NAPL/DNAPL transport model). As long as this numerical modelling is not added to the paper, the results remain too much speculative Reply: The reviewer concerns regarding absence of a quantitative model on water flow and solute transport may be addressed in this manuscript. In fact a calibrated model that is based on the measured hydraulic and chemical properties of the vadose zone has been constructed and can be add to the manuscript. Nevertheless, during the manuscript preparation we have decided to omit the model chapter from this manuscript. The reason is simply because we have found that the strength of this manuscript is in the long-term continuous data obtained from the entire flow domain and not from the model which obviously was based on the measured parameters. Moreover, we have found that the model did not add any valuable information that could not be observed directly from the measured data. The value of hypothesis based on a model vs hypothesis base on observation is a fundamental argument that requires a critical discussion before implementation.

Modeling by definition aims at extending knowledge from limited data set that may be obtained from small scale point measurements or information from the domain boundaries into larger scales or zones where the knowledge is limited. For example, vadose zone modeling often uses information from the domain boundaries at or near land surface, to understand processes taking place within the unsaturated zone where data on the dynamics of water flow and solute transport is limited. Nevertheless, the model inherently bear substantial amount of basic assumption and therefore “quantitative modeling” is by definition speculative. However, in absence of quantitative observations on the flow dynamics within the domain, as often found in vadose zone studies, the model is the only practical tools for processes quantification. Nevertheless, whenever the hydraulic or chemical characteristics within the domain can be measured continuously and provided direct indication to the dynamics of flow and transport, as demonstrated in our manuscript, then modeling is not the “sol and only” mean for quantitative analysis. It is obvious that monitoring and measurements in the unsaturated zone, sophisticated as can be, are also limited in their capability to describe the flow and transport processes (technology and method dependency). Therefore, the implications from both, the model approach and the monitoring approach are, to some extent, speculative and not presenting the “truth and nothing but the truth”. In this manuscript we used for direct and continuous measurements of hydraulic and chemical characteristics of unsaturated zone to quantification the dynamics of water flow and solute transport within the entire domain. Nevertheless, although we believe that the strength of this manuscript is in the data and hypotheses which were established on direct observation will be able to add the model chapter to this manuscript if you find it critical.

Specific comments

Comment: Line 103. Study site. Can the origin of perchlorate in the study site be identified?

Reply: The site is a former waste pond of an ammonium perchlorate factory. The origin of the perchlorate in the soil is well defined, as described in details in Gal et al. 2008, 2009.

Comment: Line 121. Heterogeneity in sedimentary vadose zone formations is omnipresent. Hence, how reliable is the single borehole to assess the lithology of the study site. Is the information of the borehole consistent with information obtained from the boreholes in the vicinity of the sampling point? Reply: In this manuscript we present the lithology and concentration as measured in a borehole that was drilled for this project in the center of the experiment site (30X10 m). Nevertheless, several other boreholes were drilled in this site and a general agreement in both lithology and concentration profiles were found (Gal et al., 2008, 2009). This has been expressed in the manuscript p. 5 line 118. Comment: Line 152. The high suspected correlation between chloride and perchlorate concentrations demonstrates that there is some natural attenuation. This is in contrast with the statement in the literature review (line 86).

Reply: The limited natural attenuation of perchlorate in the site was reported extensively in Gal et al 2008, 2009. Nevertheless we do not understand how chloride/perchlorate correlation demonstrates natural attenuation. On the contrary, perchlorate reduction should have been resulted in increased chloride/perchlorate ration as demonstrated in figure 8. It is important to note that chloride was present in the soil as described previously. It is not possible to say that the chloride originated from perchlorate reduction Gal et al 2008, 2009.

Comment: Line 198. Explain more in detail how ethanol can eliminate increased salinity. Reply: One of the most common electron donor used for perchlorate biodegradation is Sodium acetate. Therefore, application of large amounts of sodium-acetate may end-up in salinization and potentially sodification of the vadose zone.

Comment: Line 214. Specify for each infiltration pulse how much time was needed to apply the water/tracer/ethanol (hence the application rates). Also, add an estimate of the saturated hydraulic conductivity of the different layers to demonstrate that the infiltration rates stayed sufficiently below the ponding infiltration rate.

Reply: Infiltration pulses were applied through drip irrigation system with a constant drip rate of 2.2 l/h and in distribution of 0.3X0.3 m (stated in line 191 in the mnuscript). Accordingly the application rate is 0.024 m/h, which is far below the soil Ks which is ~1 m/h (loamy sand). As such the application time of each phase is derived directly from the volume divided by the discharge rate. All of which appears in chapter 3.3 Infiltration experiment and table 2. For clarification the total discharge rate (6 m3/h over the entire area) will be added to the manuscript. No ponding conditions were observed on surface and the sediment water content in the unsaturated zone remain below saturation. Due to a technical mistake during submission the water content hydrographs (figure 3 in original manuscript) was submitted without the legend and depth specification. Figure1 below includes this missing information. Note that in any case the water application time in all infiltration events was in the scale of hours (7, 14, and 42 h) compare with the variation in the vadose zone water content, as presented in figure 3, is in time scale of months.

Comment: Line 250. Significant at which statistical level?

Reply: see reply to comment p 17 of reviewer 1

Comment: Line 287. Specify exactly how the wetting front velocities are determined. We are definitely in strong transient flow conditions. Hence the wetting front velocities will vary dynamically in time.

Reply: it is obvious that an infiltration event creates field of velocities that dynamically vary in space and time. Yet, (as stated in line 284), the wetting front propagation velocity, which reflect the natural gravitational drainage across the unsaturated zone, is calculated from the wetting sequence with respect to the infiltration events on land surface. The figure below describes the wetting sequence with depth at the 3 infiltration experiments. It present the time from initiation of the infiltration event to the measured increase in water content as shown in figure 2. In addition Table 1 describes the calculated velocities to the various depths in all three experiments.

âĂČ Table 1. Velocity calculation for wetting front propagation first infiltration experiment second infiltration experiment third infiltration experiment

Depth (m) arrival time (hr) velocity (m/hr) arrival time (hr) velocity (m/hr) arrival time (hr) velocity (m/hr) 0.5 N/D N/D 5 0.10 7 0.07 2.6 20 0.13 13 0.20 16 0.16 5.5 28 0.20 25 0.22 25 0.22 8.4 40 0.21 37 0.23 33 0.25 11.2 N/D N/D N/D N/D 142 0.08

Comment: Line 290. Be more rigorous and more specific with respect to ‘flow velocities’. How are these “flow velocities” defined in a heterogeneous and time dynamic flow system? (Cf.a major concern on the need to confront such statements with those from

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a quantitative numerical model). Reply: Direct calculating of wetting front propagation velocity from the temporal variation in the vadose zone water content is a basic technique which has been described in numerous publications (Dahan et al 2007, 2008, 2009, Rimon et al 2007, 2011, all of which are cited in the manuscript). It has been further used to calibrate flow and transport models in the unsaturated zone (Turkeltaub 20014, 2015a, 2015b, 2016). As stated above, whenever high resolution hydraulic data may be obtained from the unsaturated zone then modeling is not the "sol" quantitative tool. And direct measurement of flow velocities is achievable.

Comment: Line 302. Legend incomplete. What are the different coloured curves? Where are the results of the 11 sampling units? Quid results of the control units in the top layer (0,5 and 1.3 m depths)?

Reply: The comment is absolutely right, and we are sorry for this technical mistake (see figure 2 here). Comment: Line 302. Explain more in detail the observed curves. E.g. what happens with the TDR probe at the top (I suppose) during the third infiltration event? The drainage curve looks completely different. So what happened?

Reply: We agree that it was hard to understand the wetting and drainage cycles without the legend and further explanation of the velocity calculation. We hope with our reply to previous three comments the subject is now clearer.

Comment: Line 356. This statement can't be supported. This can only be concluded if mass conservation is checked. You can have lateral flow dissipation in such system. Only, a comparison of the results with the results of a numerical mass conservative model can support such conclusions. Reply: We can hardly agree with the reviewer comment that "Only, a comparison of the results with the results of a numerical mass conservative model can support such conclusions". In this section (Lines 353-358) we describe how continuous measurement of ethanol concentration across the profile dropped to practically zero. What is it if not a direct mass conservation check; which show that the entire mas of ethanol had consumed during microbial activity? No model can give higher degree of confidence in such mas balance. Especially, when it is compared with the transport of a conservative tracer such as Br. We have dedicated a special chapter (4.4 transport and degradation) which deal with mas conservation of degradable and non degradable substance during infiltration experiment. Comment: Line 400-402. Show this in an explicit way.

Reply: Here again we present the dynamic variation in concentration of degradable (ethanol) and no degradable (Br) substance transported together in the unsaturated zone. We show how the mass of Br is conserved while the mass of ethanol is reduced in an environment that is by definition biological active. It is presented as time series of the ethanol (figure 6 in the manuscript) along time series of Br (presented as profile variations in figure 7). Accordingly we do not understand what is the meaning of more explicit way.

Comment: Line 426. Confusing legend. 1/3 -11/4 2011. Specify which data at which date exactly. Reply: Due to technical analytical problem we had to combine data from two consequent dates 1 March 2011 and 11 April 2011, which represent the ending period.

Comment: Line 451. There are other studies showing that the clay layers will have considerable impact on the vadose zone dispersion (See e.g. Javaux M. and M. Vanclooster, 2004. In situ long-term chloride transport through a layered, non-saturated subsoil.1. Data set, interpolation methodology and results. Vadose zone journal 3 : 1331-1339.).

Reply: We fully agree with the reviewer comment that a clay layer in the unsaturated zone may impact the dispersion. In fact this is something that we also found in our studies on water infiltration in layered vadose zone. Nevertheless, our statement refers to the infiltration capacity, in terms of flow velocity and fluxes. Several different and independent studies showed that the presence of the clay layer in the unsaturated zone do not limit the flow velocity (Dahan et al 2009, Rimon et al 2007, 2011, Baram et al 2012, Turkeltaub 2015). A clarification sentence will be added to the manuscript.

Comment: Line 461. This has not been shown in the paper.

Reply: The reviewer statement that the sentence “It seems that the entire column of perchlorate mass was pushed down by the percolating water toward the water table, which also resulted in an increased concentration of perchlorate in the observation well, which was located under the infiltration zone.” has not been shown in the paper is not clear. Figure 5 presents variation in perchlorate concentration profile during the infiltration experiment. It exhibit increased concentration of perchlorate in zones underlying layers of higher concentration as a response to water infiltration. This is a unequivocal indication to solute displacement.

Please also note the supplement to this comment:
http://www.hydrol-earth-syst-sci-discuss.net/hess-2016-663/hess-2016-663-AC2-supplement.pdf

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Interactive comment on Hydrol. Earth Syst. Sci. Discuss., doi:10.5194/hess-2016-663, 2017.

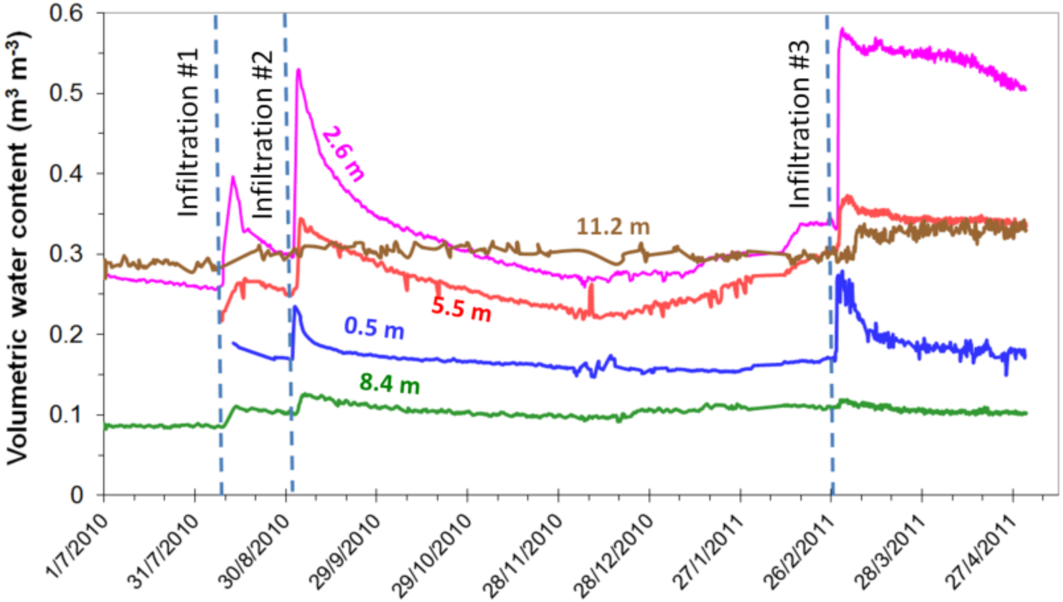

Fig. 1. Figure 1 (figure 3 in the manuscript). Temporal variations in sediment water content in the top 13 m of the vadose zone during the infiltration experiments. Dates are given as day/month/year.

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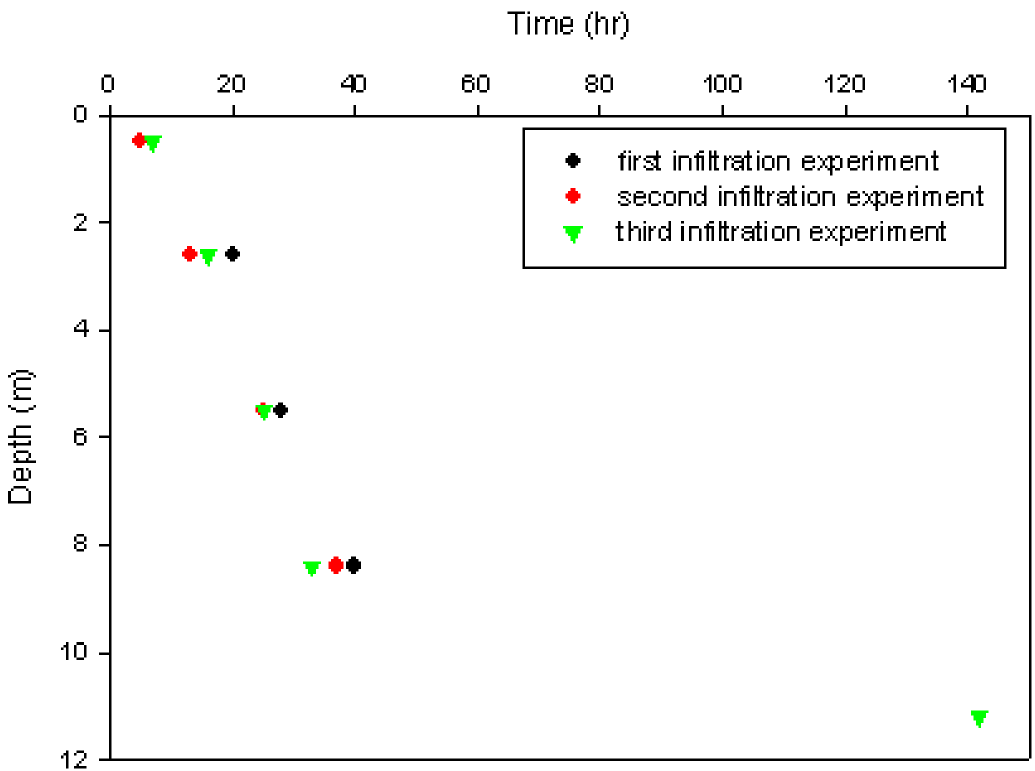

Fig. 2. Figure 2. Wetting front propagation in the upper part of the vadose zone during all three infiltration experiments, represented by the time of first measured increase in water content V.S. depth.

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