Reply to reviewer #1 comments on the manuscript:

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

We thank the reviewer for his constructive review and intend to address all of his comments. We would like to state that we are specifically encouraged by his statement "*The presented topic is of relevance for many sites worldwide, polluted with different chemicals which can be deactivated by microbial processes. The specific challenge of this approach was the location of the pollution within a deep vadose zone with complicated water flow conditions*". In our view this is the main essence of this manuscript.

General comments

Comment: Some context would be easier to understand if the order of subsections would be rearranged. For example: Section 4.3 explains why the different treatments for the experiments were chosen, because the infiltration depth was not sufficient in the beginning and the concentration of ethanol was too low during the first experiment. It would be good to have this information already in the beginning before the results of perchlorate transformation are shown and discussed. The same is true for the presentation of bromide tracer behavior (in the beginning of section 4.4) which again explains the experimental setup.

Reply: We accept the comment. A section describing the overall structure of all three experiments will be added to the beginning of the result chapter. It will present the rationale behind all experiments and gives an overview of the measurements before detailed description of the various components.

Specific comments

Comment: p. 5, l. 111: You state that perchlorate is slowly leached into the groundwater. Can you describe the behavior of this pollutant in the saturated zone? Is it reduced or only transported by groundwater flows?

Reply: Perchlorate is well known to be fairly stable in groundwater. Its natural degradation is very limited and it is highly mobile. This has been presented in several publications (See for example a review paper by Bardiya et al. 2011, a chapter in a book Coates JD, Gu B. 2006, and perchlorate mobilization in this particular site Gal et al. 2009 (all of which are cited in this manuscript). The possibility of perchlorate reduction is depend in the groundwater redox conditions. We had reported in the past that groundwater is aerobic and thus natural degradation of perchlorate is not expected (Bernstein et al., 2010). Since our manuscript focus on the vadose zone where the hydro-chemical and biological conditions are substantially different from those occurring in groundwater we rather to focus on the vadose zone and not elaborate on the saturated part beyond the limited citations in the introduction chapter.

(Bernstein, A., Adar, E., Ronen, Z., Lowag, H., Stichler, W., & Meckenstock, R. U. (2010). Quantifying RDX biodegradation in groundwater using δ 15 N isotope analysis. Journal of contaminant hydrology, 111(1), 25-35.)

Comment: p. 6, l. 147: What is the effect of these climatic conditions? Is the perchlorate only transported during the winter season and probably rises again during summer due to capillary action?

Reply: The vadose zone is very thick (~40 m) and mostly sandy. As such capillary action is not relevant and will not impact more than the bottom ~1 m. The experimental area has been covered with a sealing liner to prevent air penetration and to promote reducing conditions in the vadose zone. As such the only source of water to the subsurface in this period is the water injected to the soil with the drip irrigation system under the surface cover. Accordingly the consequence of rain water infiltration is eliminated. In addition in such thick vadose zone even seasonal temperature fluctuations are limited to the upper 2 m (Rimon et al. 2011b, cited in the manuscript). As such we believe that the climate has only limited impact on the conditions in the subsurface.

Comment: p. 11, l. 229: Please explain why no tracer was used in the second and third application.

Replay: A single slug of tracer was used in in the beginning of the first experiment. It was designed to enable tracing of the wetting front that was introduced to the subsurface during the experiment. Application the tracer in the following experiment would have result in smearing the identity of the front and masking our capability to trace the moving water. In well-defined medium such as column experiment it is possible to differ between tracers applied in different stages. Yet we tend to believe that in natural heterogeneous system where water flow may be subjected to multi flow trajectories that may be activated and deactivated according to the hydraulic condition (see Dahan et al. 2009), application of the tracer in the following experiments would be a disadvantage.

Comment: p. 12, l. 272 Can you exclude lateral fluxes of seepage water?

Reply: We cannot absolutely exclude local limited of lateral fluxes. Nevertheless, creation of lateral flow in the unsaturated zone require, by definition, generation of saturated conditions that will create positive pressure which could overcame gravitational drainage. Up to date the vadose zone monitoring system has been installed in dozens of sites with different geological and hydrological conditions (See for example Dahan et al., 2007, Dahan et al. 2008, Rimon et al 2007, 2011a, 2011b, Amiaz et al 2012 and others). In none of these sites we found evidences for creation of saturation conditions and thus creation of lateral flow in the vadose zone, even though some of the sites were under flooded conditions of high water head (Dahan et al 2007, 2008), some with geological formations which are composed of clay interbeds that could potential create some kind of hydrological barrier and lateral flow. Since we did not find any indication for lateral flow in any of the other studies where water flow in the vadose zone was monitored we tend to believe that in this particular site lateral flow, if any, was very limited. In

this discussion we ignored lateral small scale capillary flow and lateral flow in purged aquifers. Both are not relevant to this site.

Comment: p. 15, l. 326: Is the described successful reduction of perchlorate concentration the result of transport or reduction processes? Would it be a success if perchlorate is mainly transported by seepage water into deeper parts of the soil?

Comment: p. 16, l. 333: You mention mixed trends for both transformation and mobilization processes. Could you explain this conclusion more in detail?

Reply to the two comment above (p.15 and p.16): This comment emphasize the greatest challenge we faced in this project. Can we absolutely state that the reduction in perchlorate concentration that we have observed in the upper parts of the unsaturated zone are the result of bio-degradation or simple down leaching with the percolating water. Moreover, we have to investigate this question in light of the fact that the concentration of perchlorate in some deep section only increased during the infiltration experiments. Throughout the paper we have discussed the potential degradation versus leaching from different prespective. In section 4.3 we have analyzed the potential degradation of perchlorate to the availability of electron donor. Obviously under absence of available electron donor no perchlorate degradation will take place. Though we managed to introduce electron donor into the vadose zone it was limited to the top 13 m. only there we found some reduction in perchlorate. In the rest of the profile we found no increase in available electron donor and in fact we also found no reduction perchlorate concentration. On the contrary in some places the concentration only increased which is an obvious indication to perchlorate mobilization with the percolating water. Further down in the manuscript in section 4.4 we discussed the potential degradation of perchlorate versus its transport through a comparison of the ethanol migration, which was consumed, versus the tracer, Br. Here we also compared the reduction in perchlorate with the variations in concentration of the degradation by-product chloride across the unsaturated zone and found a pronounced increase in Cl/Perchlorate only in the zones where we found available electron donor. All of these indicators provided hints to the question on the degradation vs leaching.

In the second part of the first comment the reviewer ask if "it be a success if perchlorate is mainly transported by seepage water into deeper parts of the soil". This is a very important question that is the subject of several studies we are conducting now (See Avishai et al 2016. Journal of Hazardous Materials). Since we found that achieving "efficient" degrading conditions in deep vadose zone is limited and we found that perchlorate mobilization in the unsaturated zone is very high we are testing the possibility to leach the pollution down to the groundwater where it can be retrieved back for treatment on land surface.

Comment: p. 17, l. 350: Probably the relation between ethanol concentration and DOC could be shown by means of a figure and a regression curve?

Reply: As mentioned in p.17 lines 346-352, we found high correlation between ethanol and DOC. Even though ethanol is mineralized by perchlorate reducing bacteria, it may degraded first to acetate that also serve as energy for the degrading bacteria thus, DOC provide better

picture on the availability of electron donor in the soil pore water. Since it is all presented in the manuscript text we believe that adding this information in a figure is somewhat not necessary. The figure below display ethanol vs DOC in all water samples were both ethanol and DOC were measured

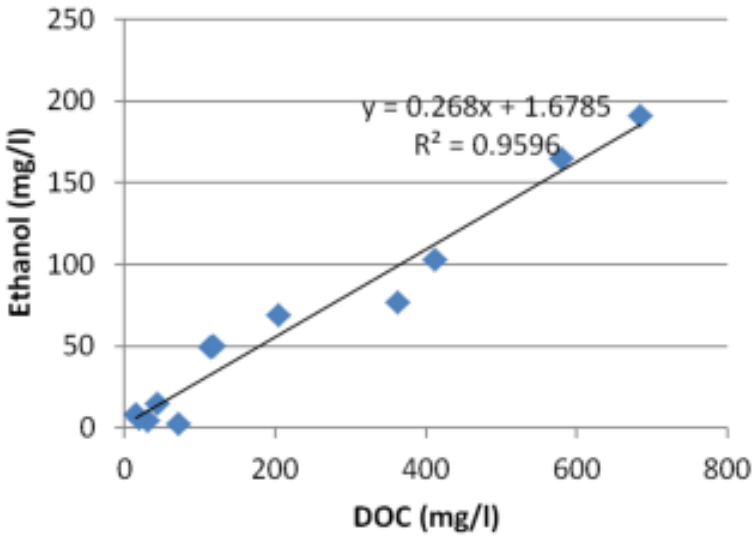

Figure 1. Ethanol VS DOC in all water samples where both were measured

Comment: p. 21, fig. 8: Is the red graph an average for data of the period 1/3-11/4 2015 (1.5 months)?

Reply: The red graph is a combination of data obtained from two consequent sampling data. Due to a technical problem that was resulted in luck of samples from one of the dates it was necessary to integrate data from these two consequent dates.

Comment: p. 22, l. 459: You end up with the conclusion that the entire column of perchlorate was pushed downwards by the infiltrating water. Thus, the problem is mainly shifted to the groundwater. Could you discuss the overall success of the presented remediation experiment against this background?

Reply: See reply to second part of comment p.15 in lines 89-95 of this document

Technical corrections

Comment: References: Bauterse et al (2000) and Stumpp et al. (2009) are not mentioned in the text

Reply: Will be corrected it in the revised manuscript

Comment: Fig. 3: the legend is missing

Reply: Will be corrected in the revised manuscript

Comment: Fig. 4/5: explain the meaning of the red arrows.

Reply: The red arrows emphasize the variation in perchlorate concentration in time. In Figure 4 it describe perchlorate reduction in the upper 13 m while in figure 5 the arrow emphasize the increase in perchlorate concentration with time in the deeper section of the vadose zone. Elaboration on the meaning of the arrows will be added to the figure captions.