

The authors present a novel in-situ, bioremediation method for the treatment of polluted waters from the unsaturated zone and from the groundwater. The method relies on a cyclic process in which contaminated groundwater is pumped to the surface and subsequently, it is injected back into the topsoil (after the addition of an electron donor). The topsoil area acts like a bioreactor in which contaminants are removed by microbial degradation. Cleaned water is then propagating through the unsaturated zone to greater depths and pushing down further contaminated water fractions to the groundwater where it is then again pumped back to the surface, closing the cyclic process. In this way, all contaminants are gradually removed.

The authors present experimental results on the removing of perchlorate and other contaminants at a field site over several months. They calibrate and validate the HYDRUS 1D model on experimental data (i) to capture the observed breakthrough behaviour of perchlorate over the vertical extent of the unsaturated zone, (ii) to quantify the perchlorate breakthrough into the groundwater aquifer and (iii) to predict the time scale for a complete removal of perchlorate and all other contaminants at the specific field site.

I think that such technical methods for removing contaminants out of the subsoil are a crucial tool for protecting water resources and ecosystem. Hence, the present study is relevant for the readers of this journal. In general, I could quite easily follow the descriptions and common thread of the study as the structure and build-up of the text was clear to me. However, despite the relevance of the topic and the good readability of the study, I would recommend some moderate to major revisions due to the following general and specific comments.

### **General comments**

1. For me it remains quite unclear which of the presented experimental results/data have been published before and which are completely new in this study. The reader can get the impression that the former is true for all observed data and that “only” the simulation part is new (until reading the Conclusions). At the beginning of the study, please make explicitly clear in a short passage which experimental results are new and which have been published before, and maybe also stress the differences of the current experiment setups compared to the previous experiments at that site.
2. For the simulation part, it would be interesting to see uncertainty ranges and sensitivities of the simulation results to different soil hydraulic parameterizations (at least for the calibration), as models like HYDRUS have several degrees of freedom and are quite sensitive to the soil hydraulic functions.
3. In the discussion, the authors may additionally comment on:
  - (i) The influence of preferential leaching (e.g. in macropores) of contaminants at that site. Don't you have any evidence for preferential leaching and a bypassing of the bioreactor zone through heterogeneous soil structures, because you obviously use an one-porosity approach for the HYDRUS simulations assuming well-mixed conditions? I think especially for the presented in-situ remediation method with biodegradation in the shallow unsaturated soil zone the assessment of the influence of preferential leaching is crucial, also for the general transferability and applicability of the method at other sites.

- (ii) General differences of the presented method to other, commonly used methods in terms of, e.g. the scope, costs, environmental sustainability, risks and challenges.

### **Specific comments**

1. P. 1, abstract: You use some abbreviations directly at the beginning of your study without giving their entire name once when using the terms for the first time. Please also check this issue throughout the study as it occurs more often (e.g. in the methods section).
2. P. 1, l. 32: I was wondering if there is one classical method that is most commonly used for remediation to which the presented in-situ bioremediation method and the specific perchlorate results of this study can be directly compared (cf. my last general comment).
3. P. 2, l. 36-42: How relevant is the contamination of ecosystems by perchlorate. Do you have any specific numbers on the magnitude of perchlorate contamination on a global or national scale?
4. P.3, Figure 1: The quality of the legend picture is poor and hardly readable. Please try to increase the readability. Also, the left part of the legend is not further defined here but is only described later in the text. It would be easier for the reader if these 5 different soil layers were already properly defined here in Figure 1.
5. P. 4, l. 115: Which suction pressure/head was used to sample the pore water in different depths? Was it a constant pressure or variable depending on the soil water content? Please provide more information on that.
6. P. 5, Figure 2: I really like this schematic sketch, it provides a good overview.
7. P. 6, l. 147: In this case it must be “advection-dispersion-equation (ADE)”.
8. P. 6, l. 165: How was  $v$  determined? Measured, calibrated? In general, what are the used values for  $v$ ,  $D$ ,  $\tau_w$ ? Maybe you can show a Table with all parameters values used for the simulations, at least in the Appendix.
9. P. 7, l. 196: Can you please provide some more information about the determination of hydraulic properties of the different soil layers? In the previous experiments at this site over the last years, were there no actual measurements of the hydraulic properties (and the generation of soil water retention and hydraulic conductivity curves) of soil samples in different depths?
10. P. 7, l. 207: This would imply a steady-state condition. Is there any experimental evidence for this assumption? And in the next line, how low is this regional gradient that reasons the closed domain assumption?
11. P. 8, l. 217: In line with my specific comment #8, can you please provide an overview of the used parameter values of  $A_{ex}$ ,  $Z_{box}$ ,  $V_{total}$ ?

12. P. 8, l. 221: I think you mean “The initial mass of perchlorate is...”.
13. P. 8, Eqs. 7 and 8: The unit conversion factor must be “1000 L / 1 m<sup>3</sup>” for consistency.
14. P. 9, l. 239: Can you here please give a short explanation or suggestion why the diffusivity in the last two layers is so high compared to the other layers above?
15. P. 10, l. 263: Why even showing and using the observed values in 17 m depth for simulation, when you think that this sampling point does not generate reliable measurements and not capturing the real conditions in this depth?
16. P. 10, Table 1: The unit of  $\lambda$  is missing.
17. P. 12, l. 310-313: This explanation is not completely clear to me. Can you please provide some more information on this modelling procedure in two phases? How long are the respective phases? And how do you subdivide Layer 1 from Table 1 (0-2m) into the three sublayers of Table 3 between 0-40 m?
18. P. 13, Table 3: Do you assume  $\mu$  as the first-order degradation rate coefficient for the water phase? Thus, do you only assume degradation taking place in the water phase, as you do not say anything about parameterizing sorption? Does perchlorate not adsorb at all to soil particles?
19. P. 14, l. 363: I cannot find a depth of 0.05 m in Figure 4. Please revise.
20. P. 15, Figure 7: Labels of x-axis are missing.
21. P. 18, l. 446: What does “minimal costs” actually mean? What would a continuous treatment over 900 days cost and what is the difference to common methods?