**General comments**

**Comment 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.
**Reply to comment 1:**
The comment is accepted, and the manuscript was revised accordingly. (The only results that were previously published are the trace experiment).
Lines 104: " The study includes a new treatment experiment conducted for nine months from October 2019 to June 2020." Line 131: " The current remediation experiment is presented for the first time "
Lines 143-145:
"In addition to the new result from the long-term experiment (section 2.2), former data from a trace experiment was used for the assessment of the water flow in the unsaturated zone. This experiment was conducted in 2015 in the same research field and previously presented by Levakov et al. (2019)."

The differences between the current and former experiments are mentioned in the conclusions section in lines 425-429. Nevertheless, an additional explanation was added to the manuscript: lines 131-134: " The current remediation experiment is presented for the first time, and the applied conditions in it were based on previous conclusions from the research site. The recent improvements to the experiment setup included the addition of a buffer to the system and the use of lower concentrations of electron donors in order to maintain optimal pH values for the degradation process."

**Comment 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.
**Reply to comment 2:** We accepted the reviewer's suggestion, and the Morris method sensitivity analysis has been applied (Morris, 1991). In this method, the parameters are modified one-at-a-time (OAT) and sensitivity is estimated as the partial derivative of the change in model output for a given change in a single input parameter (Perzan et al., 2021). We used the SAFE Matlab code provided by Pianosi et al. (2015). Furthermore, the sensitive parameters were calibrated following the method presented by Perzan et al. (2021), where uncertainty and optimal values are calculated according to the behavioral simulations (by achieving evaluation goals). Note that the 'Methods' and the 'Results and 'discussion' sections have been revised (Lines 214-250;282-316; Tables 1 and 2; Figures 3-6).

**Comment 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.
**Reply to comment 3(i):** Preferential flow in the unsaturated zone is expected. Moreover, our monitoring technology which measures water flow and solute transport in the unsaturated zone provides a clear indication for preferential flow as it provides information from multiple independent monitoring points across the unsaturated zone, where each point is located under a different unsaturated column. Quantifying the extent of the preferential flow both as reflected from the field

measurement and through the model is not straightforward. Yet, in this study, which was conducted in unconsolidated sandy and clay sediment, we could clearly see that the infiltrating water does not have a significant bypass of the bioactive zone. The indications come from the fact that multiple monitoring points at different depths under different soil profiles where all show reasonably coherent results. This enabled us to simplify the model while accounting for single soil properties in the different layers.

(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.

**Reply to comment 3(ii):**

The comment is accepted, and the manuscript was revised accordingly.

The common methods for perchlorate treatment were elaborated in the introduction: lines 58-68:

"Perchlorate contaminations are treated around the world according to the nature of the contaminated site. In the case of polluted soil and deep unsaturated zone, in-situ techniques are used mainly based on biodegradation by native microbial populations (Bardiya and Bae, 2011; Coates and Achenbach, 2004; Xu et al., 2003). For example, injections of carbon-rich solutions (Frankel and Owsianiak, 2005) or gases (Cai et al., 2010; Evans et al., 2011) supplied electron donors for the microbial reduction processes directly to the soil at different depths up to 10m. As opposed to those methods, the current research site is characterized with a deep unsaturated cross-section (40m) while the major mass of perchlorate is located in the deep layers (17-36 m) (Dahan et al., 2017; Gal et al., 2009; Levakov et al., 2019). For pollution in the deep unsaturated zone, flushing is a common way to treat deep and inaccessible layers by injecting clean water to the site, displacing the pollutions to the groundwater, and collecting it back to the surface for further treatment (L. Liu et al., 2018; Luciano et al., 2013; Wellman et al., 2011). Usually, the subsequent treatment includes ex-situ methods outside the contaminated site (Guo et al., 2013; Høisæter et al., 2021)"

And the comparison to our technique was added to the conclusions: lines 528-536:

"Usually, different in-situ biodegradation techniques are used for contaminated soils up to a deep of 10m, with lower perchlorate concentrations (Bardiya and Bae, 2011; Coates and Achenbach, 2004; Xu et al., 2003, Frankel and Owsianiak, 2005, Cai et al., 2010; Evans et al., 2011). An additional approach is an excavation of the contaminated soil followed by an ex-situ treatment outside the site. For deeper contamination, the common treatment approach involves an injection of clean water, displacing the pollution towards the water table, and collecting it back to the surface for further treatment ex-situly. All ex-situ approaches require relatively high costs and complicated operations during the transport of the contaminants and the establishment of the treatment facilities. Unlike those methods, the proposed approach treats the contamination in-situly and simultaneously for the soil, the unsaturated zone and the groundwater with extremely high perchlorate concentrations in a relatively simple and affordable manner."

**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).

**Reply:** The comment is accepted, and the manuscript was revised accordingly. Lines 9,16,18,35, and 92.

**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).

**Reply:** The comment is accepted, see reply to comment 3(ii).

**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?

**Reply:** The comment is accepted, and the introduction section was revised accordingly. Lines 39-41:
" Since the late 1990s, the recognition of perchlorate contamination was grown worldwide, especially in the USA, Canada, China, and west Europe with thousands of studies examining perchlorate contaminations in different water bodies, soils, and food products (Cao et al., 2019). "

**4**. P.3, Figure 1: The quality of the legend picture is poor and hardy 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.
**Reply:** the comment is accepted. The figure was updated and the caption was revised. Lines 101-102:
"**Figure 1. Perchlorate distribution across the unsaturated zone (left). Lithological profile (right)*, and division of the profile into five different layers as used for the Hydrus 1D model (middle).*"**

**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.
**Reply:** The technical information on the vadose zone monitoring system was published previously with an accurate description of the system's operation. In order to keep the manuscript relevant and concise we try to avoid excessive details. Nevertheless, we added a reference for more detailed information. Lines 128-129:
" For more detailed information on the vadose zone monitoring system see Dahan et al., 2009. "

**6**. P. 5, Figure 2: I really like this schematic sketch, it provides a good overview.
**Reply:** Thank you

**7**. P. 6, l. 147: In this case it must be "advection-dispersion-equation (ADE)".
**Reply:** The comment is accepted, and the term was revised. Line 163.

**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.
**Reply:** The *v* is the porewater velocity and it is assumed to be equal to the ratio between the calculated Darcian flux (calculated by the Richards equation) and the water content. Note that these values are calculated with the numerical model integrated in Hydrus 1D code (we add an explanation to the text, lines 185-186). The calculations of the Richards equation and the Advection-Dispersion equations are coupled. Similarly goes for the tortuosity ($\tau_w = \frac{\theta^{7/3}}{\theta_s{}^2}$), which changes according to the water state of the soil (see line 183). As described in equation 5 (line 181), the hydrodynamic dispersion coefficient (D) is a function of Darcian flux (or porewater velocity) (Richards equation), tortuosity, longitudinal dispersivity (λ), which is handled as a fitting parameter (lines 184-185) and $D_w$, which is the diffusion coefficient that accounts for the Brownian movement. The $D_w$ was obtained from literature, 1.55 x $10^{-4}$ $m^2$/day (lines 184-186).

**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?
**Reply:** We agree with the reviewer's comment and further explanation regarding the determination of the hydraulic parameters is provided in the text (Lines 214-250).

**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?
**Reply:** The situation in the groundwater was simplified by a steady-state condition in order to provide a picture regarding the influence of the treatment on the groundwater pollution. Although there is horizontal flow in the aquifer, it considers low especially compared to the pumping/injecting rate from/to the aquifer. Moreover, we clarified this inaccuracy in lines 391-394:
" It is important to mention that this mass balance does not consider the dispersion and advection in the aquifer, and therefore, a specific model needs to be run after setting the total components of the system, such as the current surrounding wells and local hydraulic heads. Nevertheless, it provides a rough estimation of the situation in the groundwater."
Additional clarification was added to the methods section. Lines 253-254:
" The calculation was simplified by a steady-state condition in order to provide a rough estimation for the contamination in the groundwater "

**11**. P. 8, l. 217: In line with my specific comment #8, can you please provide an overview of the used parameter values of $A_{ex}$, $Z_{box}$, $V_{total}$?
**Reply:** The comment is accepted, and the terms were elaborated. lines 267-269:
" The total volume ($V_{total}$) was calculated by multiplying the penetration area of the experiment, $A_{ex}$ ($m^2$) =240$m^2$ (see section 2.2) by the depth of the aquifer section used in the model, $Z_{box}$ (m) =10m (determined as the boundary of the model); "

**12.** P. 8, l. 221: I think you mean "The initial mass of perchlorate is…".
**Reply:** The comment is accepted, and the terms were changed. Line 271: " The initial mass of perchlorate is…"

**13.** P. 8, Eqs. 7 and 8: The unit conversion factor must be "1000 L / 1 $m^3$" for consistency.
**Reply:** the other reviewer asks to delete the unit conversion from the equation. Therefore, Eqs 10-11 were revised.

**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?
**Reply:** According to the sensitivity analysis that was implemented in the revised manuscript. It appears that the λ parameters were not sensitive. Therefore, the default value applied by the Hydrus code was kept (i.e., 10 cm; Tables 1 and 2).

**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?
**Reply:** The vadose zone monitoring system in our research field provided information on 40m of unsaturated zone, and as in any unsaturated zone the level of homogeneity and uncertainty is hard to estimate. Nevertheless, we presented the results in order to reflect the heterogeneity of the cross-section and the difficulty in evaluating the flow and solutes transport process. It is important to clarify that the data from 17m depth was not used for calibrating the model but to assess the fitting of the calibrated model to the observation in the field. And for the scientific integrity we decided to keep those unperfect results.

**16.** P. 10, Table 1: The unit of $\lambda$ is missing.
**Reply:** Table 1 has been revised. The λ parameter is in cm.

**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?
**Reply:** We apologize for the typing mistake in Table 3, the depth of the third layer should be between 1.3 and 2 m thickness. Table 3 was revised accordingly. Furthermore, we added more information regarding the modelling procedure in two phases (lines 387,389-391). Note that the first phase was running for 55 days, and the second phase extended over 233 days.

**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?
**Reply:** Perchlorate does not adsorb to soil particles as mentioned in line 37: " Perchlorate's high solubility and low sorptivity to soil particles facilitate its distribution in the subsurface "

**19.** P. 14, l. 363: I cannot find a depth of 0.05 m in Figure 4. Please revise
**Reply:** The comment is accepted and we removed reference to the figure.

**20.** P. 15, Figure 7: Labels of x-axis are missing.
**Reply:** Figure 7 has been revised accordingly**.**

**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
**Reply:**
Compared to different common methods, which usually based on ex-situ treatments (such as external reactors, soil excavation etc.), submersible pump and basic irrigation equipment are the only technical requirement for our proposed treatment. We clarified this point in lines 533-536:
" All ex-situ approaches require relatively high costs and complicated operations during the transport of the contaminants and the establishment of the treatment facilities. Unlike those methods, the proposed approach treats the contamination in-situly and simultaneously for the soil, the unsaturated zone and the groundwater with extremely high perchlorate concentrations in a relatively simple and affordable manner."