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Supplement of

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

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Vadose Zone Monitoring system (VMS)

The need for real-time information on the quality of percolating water in the unsaturated zone led to the development of a vadose-zone monitoring system (VMS). The VMS is designed to provide in-situ continuous measurements of the hydraulic and chemical properties of the entire vadose zone, from land surface to the water table (Dahan et al., 2009). The monitoring system is composed of a flexible sleeve made of thin urethane liner, hosting several customized FTDR probes, for water content tracking (Dahan et al., 2008; Rimon et al., 2007), and vadose zone sampling ports (VSPs) for sediments pore-water sampling (Dahan et al., 2014; Rimon et al., 2011; Turkeltaub et al., 2016) (Figure 1).

Accordingly information obtained by the VMS is used for direct measurements of flow velocities, solute transport and chemical transformation of the percolating water, from land surface to the water table. Up-to-date, the system has been successfully implemented in several studies on water flow and contaminant transport in various types of hydrogeological conditions including: (a) rainwater infiltration and groundwater recharge (Rimon et al., 2007; Turkeltaub et al., 2015a), (b) floodwater infiltration in arid lands (Amiaz et al., 2011; Dahan et al., 2008), and (c) agricultural impact on groundwater quality (Baram et al., 2012; Dahan et al., 2014; Turkeltaub et al., 2014, 2015b, 2016). In addition, transient data on the temporal variations in vadose zone water content and chemical composition of the sediment pore water enable calibration and validation of flow and transport models to the actual dynamic characteristics of the vadose zone (Turkeltaub et al., 2014, 2015a, 2015b, 2016).

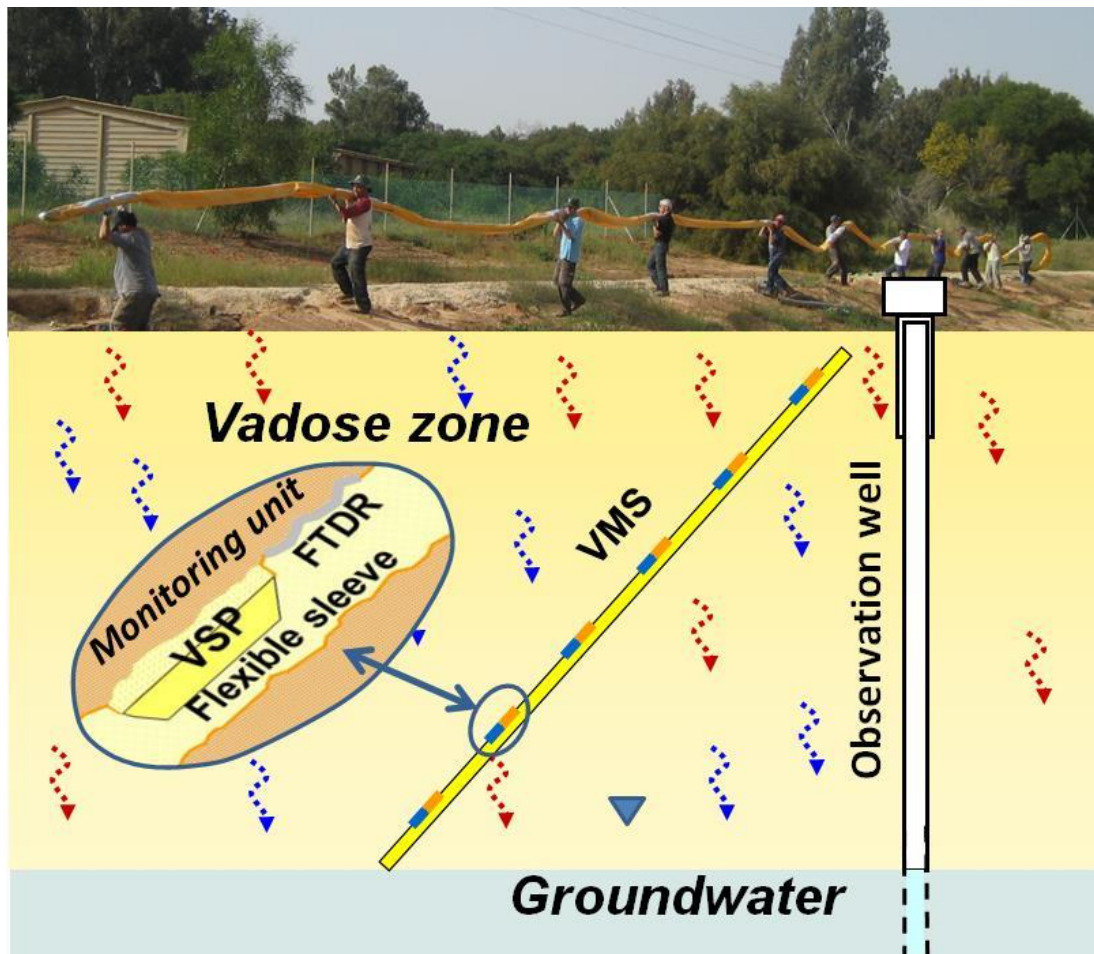


Figure 1. Schematic illustration of a vadose zone monitoring system (VMS) with its monitoring and a photo of a VMS carried on for installation.

Installation: The monitoring systems are installed in uncased, small-diameter (~ 0.15 m), slanted boreholes (55° to the horizon). The FTDR probes and VSPs are aligned along the borehole's upper side wall facing the undisturbed sediment column, which extended from the probe location on the borehole wall to the land surface. Immediately after inserting the sleeve into the borehole, it is filled with non-shrinking cement grout. The hydrostatic pressure generated by the filling material within the sleeve causes its expansion and ensures that it fills the entire void of the borehole. As a result, the FTDR probes and VSPs are pushed against the borehole's upper side wall with sufficient force to achieve good contact with the sediments. The combination of flexible sleeve with high density filling material ensures tight sealing of the borehole void and elimination of potential preferential flow and cross contamination along the sleeve.

Flexible TDR (FTDR): the FTDR probes are designed to provide information on the infiltrating water via temporal variations in the vadose zone water content. The FTDR probes are made of flexible waveguides attached to a flexible sleeve. Filling the sleeve with the cement grout causes its expansion and attachment of the flexible waveguides to borehole walls. The flexibility of the waveguides enables full contact in spite of the roughness, undulations, and diameter irregularities of the uncased borehole. The inner side of FTDR waveguides is shielded with a 3 cm thick dielectric insulation to prevent potential impact of the cement on water content measurement. The FTDR operates with Campbell Scientific data acquisition and logging instruments including TDR100, SDM50X and AM16/32 multiplexers, and CR10X data loggers. All waveguides were connected to an acquisition system using a high quality RG-58 coax cable (shielding factor >95%). Due to the known effect of cable length on TDR waveform, a linear relationship between apparent and cable length is implemented and the cable length correction are established (Rimon et al., 2007).

$$\text{Corrected La/L} = \text{measured La/L} - \text{cable L} \times 0.016$$

Finally, a calibration curve for volumetric water content measurements by FTDR probes was established for several different soils (Rimon et al., 2007). The calibration showed a linear correlation between the TDR signals (the calculated apparent length [La] divided by the real length [L]) and the volumetric water content (Figure 2). The FTDR is a patented technology (Flexible Probe for Measuring Moisture Content in Soil, USA, Patent # 6,956,381).

Vadose one pore-water sampling port (VSP): The VSP enables frequent sampling of the vadose zone pore-water. The physical mechanism that allows sampling of the vadose zone pore-water by the VSP is based on creating hydraulic continuity between the sediment pore-water and the sampling cell, similar to standard tensiometer or suction cups. Hydraulic continuity between the VSP and the rough sediments on the borehole sidewall is achieved through an unconsolidated agent made of flexible quartz powder. The VSP is operated through a set of small-diameter access pipes (i.d. 1>mm) reaching a control panel on surface. Once hydraulic continuity between the sediment pore-water and the VSP is achieved, low pressure (vacuum) is applied to the sampling cell to draw the sediment pore water to the sampling cell. The water sample is then retrieved to land surface using pressurized gas (N₂). Finally, the sampling efficiency, as expressed by the water flux into the sampling cell, is controlled by the

sediment water content. The VSP is a patented technology (Deep vadose zone sampling ports, US 12/222,069; EP 07706061.4; IL 193126).

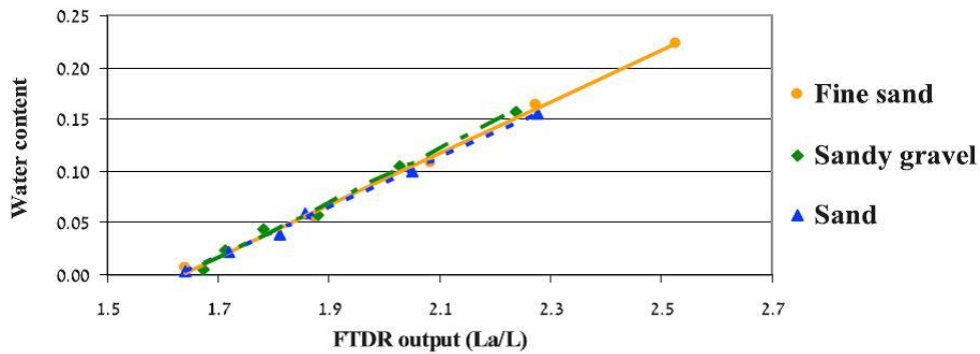


Figure 2. Calibration curve for water content measurements by FTDR probes for three different soils. FTDR output presented as apparent length/actual rod length [Rimon et al., 2007].

Calculation of percolation velocity

Direct calculating of wetting front propagation velocity from the temporal variation in the vadose zone water content, as measured by the VMS, has been described in numerous publications (Dahan et al., 2007, 2008, 2009, Rimon et al., 2007, 2011). It has been further used to calibrate flow and transport models in the unsaturated zone (Turkeltaub et al., 2014, 2015a, 2015b, 2016). The wetting front propagation velocity is calculated directly from the wetting sequence with respect to the infiltration events on land surface (Figure 3). It presents the time lag between initiation of the infiltration event on land surface to the measured increase in water content at various depths. A summary of calculated velocities to the various depths in all three experiments is presented in table 1.

Table 1. Calculated percolation velocities for the three infiltration experiments

| Depth (m) | Experiment # 1 | | Experiment # 2 | | Experiment #3 | |
|-----------|-------------------|-----------------|-------------------|-----------------|-------------------|-----------------|
| | 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 |

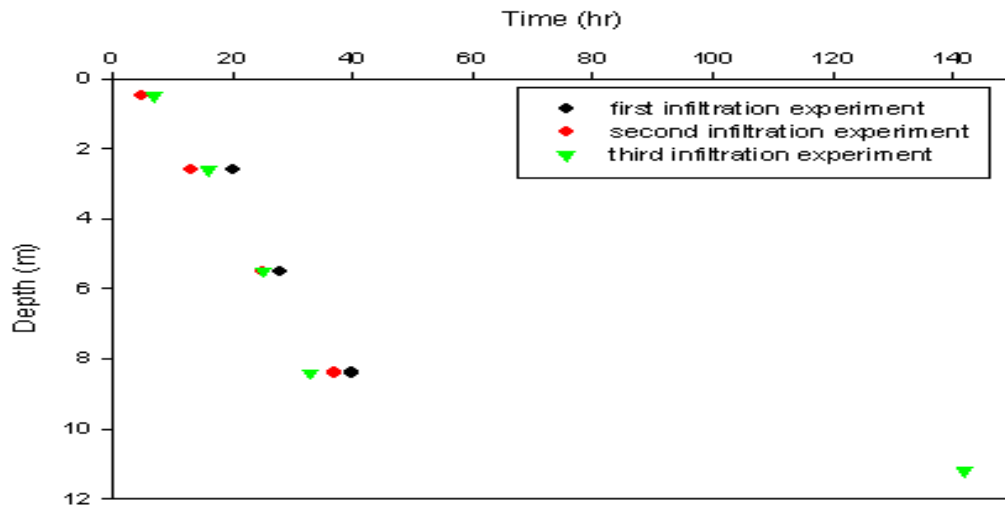


Figure 3. Wetting front propagation in the upper part of the vadose zone during all three infiltration experiments, represented by the time of first measured increase in water content V.S. depth.

Flow model

A flow and transport model was developed for the unsaturated zone at the site. The model was calibrated and validated on the basis of dynamic variations in water content and bromide concentration across the unsaturated zone. The model was developed using HYDRUS 1D. Though the data reflects multiple parallel vertical profiles, which is the outcome of the slanted installation, the 1D model results shows a relatively good fit to the data (Figures 4 and 5). Apparently the model does not provide additional insight on the flow and transport processes which could not be observed directly from the measured data. Therefore the model main results are presented in the supplementary material and not in the manuscript. Further information on calibration and validation of flow and transport models to dynamic data from deep vadose zone may be found in Turkeltaub et al., 2014, 2015a, 2015b, 2016 .

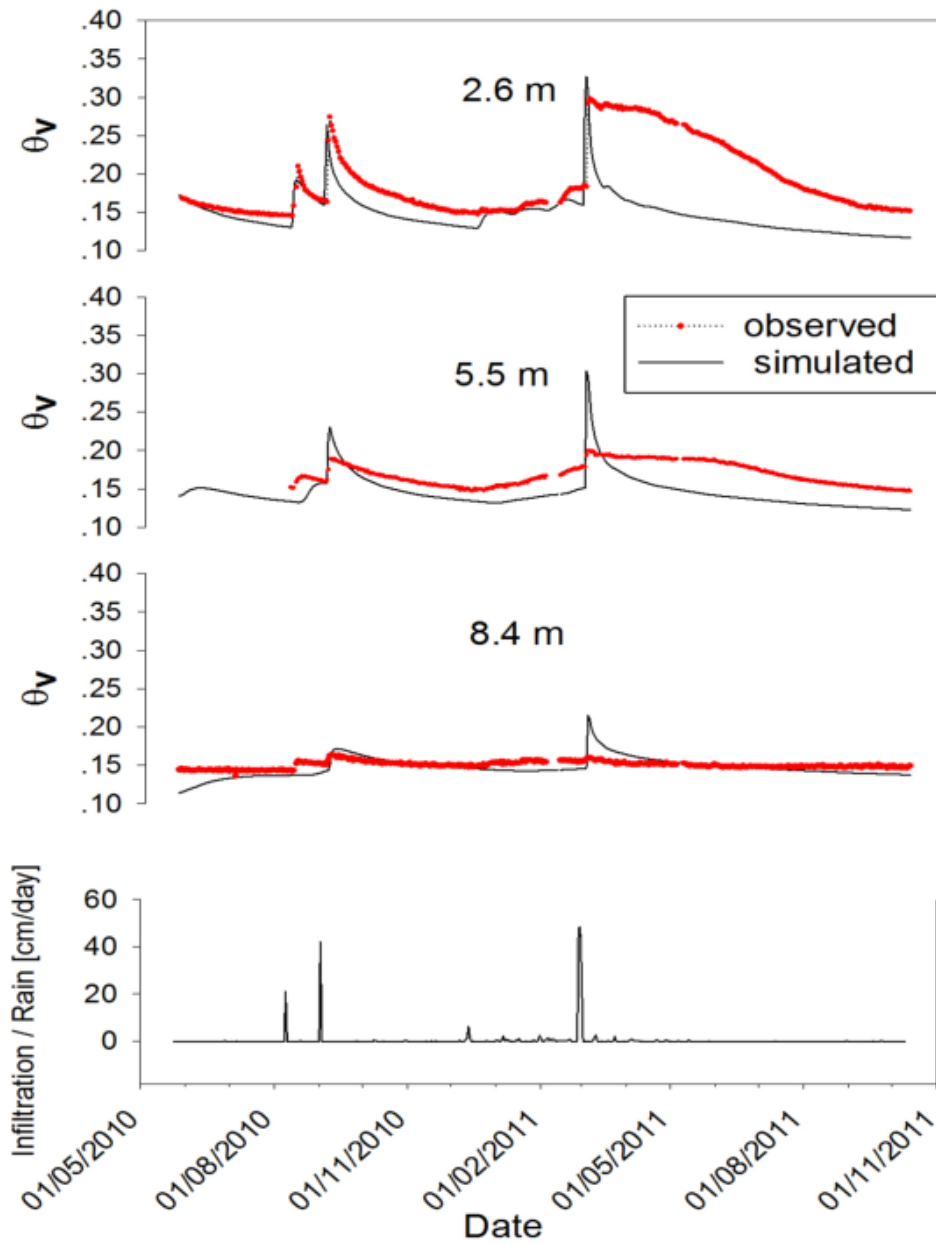


Figure 4. Measured Vs. modeled water content at various depth during infiltration experiments.

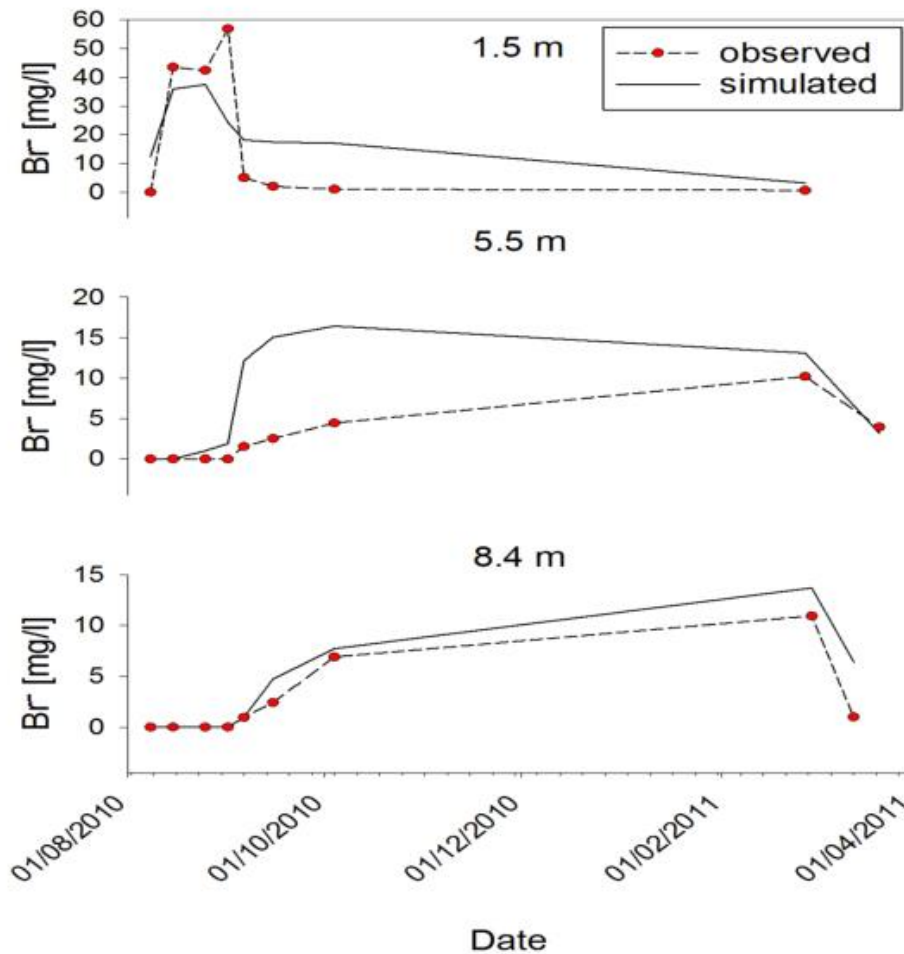


Figure 5. Measured Vs. modeled Bromide concentration at various depth during infiltration experiments.

Correlation between DOC and ethanol concentration in soil water samples

High correlation between ethanol and DOC concentration was found in water samples obtained by the VMS across the unsaturated zone (Figure 6). Even though ethanol is mineralized by perchlorate reducing bacteria, it may degraded first to acetate that also serve as energy source for the degrading bacteria thus, DOC provide better picture on the availability of electron donor in the soil pore water.

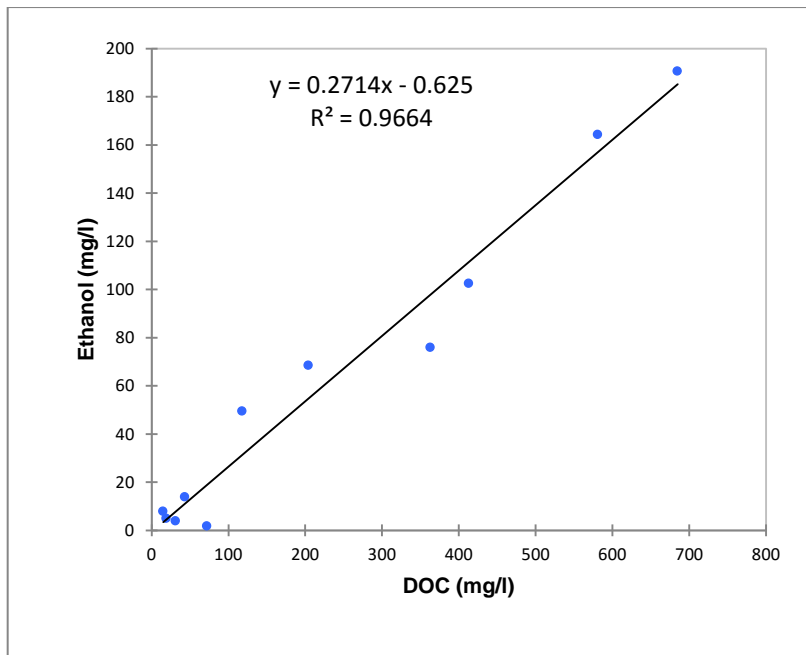


Figure 6. Ethanol VS DOC in all water samples where both were measured

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