

On the role of operational dynamics in biogeochemical efficiency of a soil aquifer treatment system

Shany Ben Moshe¹, Noam Weisbrod², Felix Barquero³, Jana Sallwey³, Ofri Orgad², and Alex Furman¹

¹Technion - Israel Institute of Technology, Civil and Environmental Engineering, Haifa 32000, Israel

²The Zuckerberg Institute for Water Research, Blaustein Institutes for Desert Research, Ben Gurion University of the Negev, Israel

³Institute for Groundwater Management, Technische Universität Dresden, Dresden, Germany

Correspondence: Shany Ben Moshe (Benmoshe.shany@gmail.com)

Abstract. Sustainable irrigation with treated wastewater (TWW) is a promising solution for water scarcity in arid and semi-arid regions. Soil aquifer treatment (SAT) provides a solution for both the need for tertiary treatment and seasonal storage of wastewater. Stresses over land use and the need to control the obtained water quality makes the optimization of SAT of great importance. This study looks into the influence of SAT systems' operational dynamics (i.e. flooding and drying periods) as well as some aspects of the inflow biochemical composition on their biogeochemical state and the ultimate outflow quality. A series of four long-column experiments was conducted, aiming to examine the effect of different flooding/drying period ratios on dissolved oxygen (DO) concentrations, oxidation-reduction potential (ORP) and outflow composition. Flooding periods were kept constant at 60 minutes for all experiments while drying periods (DP) were 2.5 and 4 times the duration of the flooding periods. Our results show that the longer DP had a significant advantage over the shorter periods in terms of DO concentrations and ORP in the upper parts of the column as well as in the deeper parts, which indicates that larger volumes of the profile were able to maintain aerobic conditions. DO concentrations in the deeper parts of the column stabilized at ~3-4 mg/L for the longer DP compared to ~1-2 mg/L for the shorter DP. This advantage was also evident in outflow composition that showed significantly lower concentrations of $NH_4^+ - N$, DOC and TKN for the longer DP (~ 0.03, ~ 1.65 and ~ 0.62 mg/L respectively) compared to the shorter DP (~ 0.5, ~ 4.4 and ~ 3.8 mg/L, respectively). Comparing experimental ORP values in response to different DP to field measurements obtained in one of the SAT ponds of the SHAFDAN, Israel, we found that despite the major scale differences between the experimental 1D system and the field 3D conditions, ORP trends in response to changes in DP, qualitatively match. We conclude that longer DP not only ensure oxidizing conditions close to the surface, but also enlarge the active (oxidizing) region of the SAT. While those results still need to be verified in full scale, they suggest that SAT can be treated as a pseudo-reactor that to a great extent could be manipulated hydraulically to achieve the desired water quality while increasing the recharge volumes.

1 Introduction

Water shortage in arid and semi-arid regions leads to great difficulties sustaining local agriculture which have many economical as well as social and environmental implications (Garcia et al., 2014). The use of treated wastewater (TWW) for irrigation is widely accepted as one of the means to reduce agricultural water scarcity (Negewo et al., 2011). While biological treatments like activated sludge are highly efficient in terms of pollutant removal, the TWW they generate usually does not meet regulatory standards for unrestricted crop irrigation (Tanji et al., 1997) or poses a sustainability question mark on their practice (Assouline et al., 2016). For example, in the Israeli Dan region wastewater treatment plant (the SHAFDAN site), following the activated sludge secondary treatment, TWW have low to moderate organic load. Dissolved Organic Carbon (DOC) is ~ 10.8 mg/L, $NH_4^+ - N$ is ~ 4.2 mg/L and organic nitrogen is ~ 1.25 mg/L (Icekson et al., 2011). However, for the TWW to meet regulatory standards for unlimited irrigation, further water quality enhancement is required (Shuval et al., 1986). Soil aquifer treatment (SAT) may be the supplementary treatment component for conventional (secondary) WW treatment, that is needed to meet regulations and sustainability. These systems involve clusters of infiltration ponds through which TWW is infiltrated through the vadose zone, into the aquifer in cycles of flooding and drying. This form of operation (i.e. flooding and drying cycles) is important for both sustaining infiltration rates (Ganot et al., 2017) and rates of microbially driven oxidation and reduction processes. In the ponds and subsequently in the unsaturated zone, residual dissolved organic carbon (DOC) and other nutrients (like organic and inorganic nitrogen species) are involved in biogeochemical processes (such as adsorption to the soil minerals, consumption by bacteria etc.) which result in further decrease in the TWW's organic load and overall improved chemical composition (Bouwer et al., 1991; Amy et al., 2007).

In Israel, ~ 150 million m^3 of wastewater (WW) are treated each year in the SHAFDAN facility (Icekson et al., 2011). After the SAT process, the TWW is transported to the south of Israel and is used by farmers for crop irrigation (Idelovitch et al., 2003). With the constant rise in population, the site is often not able to treat all the WW it receives, which results in conventionally treated WW being discarded to the local streams and the Mediterranean sea. Clearly, the SAT component is the bottleneck for full utilization of TWW. The SAT mechanism relies on the various biogeochemical processes that take place during TWW infiltration. These processes begin even before the TWW reach the unsaturated zone. Goren et al. (2014) described the variability in carbon and nitrogen species through the hours of the day and in different seasons in a field study conducted in the SHAFDAN site. They found that the chemical composition of the TWW in the infiltration ponds responded to the day and night cycles. For example, during daytime, dissolved oxygen (DO) concentrations increase due to photosynthesis, reaching a maximum in the late afternoon. As a result, TWW that are infiltrated during the day are significantly more oxidized compared to TWW that is infiltrated during the night, affecting the redox state of the soil profile and thus impact rates of oxidation reactions. Once the TWW reaches the unsaturated zone, oxygen concentrations become a limiting factor that affects the efficiency of some key biogeochemical processes that are crucial for the enhancement of the TWW quality. The role of oxygen as a limiting factor in SAT systems is especially prominent in the deeper areas of the vadose zone where natural aeration during the drying periods (DP) is limited. DOC and nitrogen species' degradation during infiltration depend heavily on processes like aerobic bacteria respiration and nitrification for which DO concentrations are of great importance (Goren et al.,

55 2014). Mienis et al. (2018) studied nitrogen behavior under the SHAFDAN's infiltration ponds through 40 years of operation .
In their study, concentrations of NH_4^+ , Total Kjeldahl Nitrogen (TKN; The total concentration of organic nitrogen and NH_4^+)
and oxidized nitrogen species (NO_2^- , NO_3^- , NO) were monitored, using data from two observation wells, screening different
depths of the vadose zone. They found that removal of up to $\sim 75\%$ of the total nitrogen occurred in the upper parts of the
soil profile (up to ~ 70 cm below ground surface) while the deeper parts had a smaller contribution. This observation led them
60 to the conclusion that inflow concentrations of more than 8.3 mg/L of $NH_4^+ - N$ will result in a decrease in reclaimed water
quality due to NH_4^+ and organic nitrogen leakage into the aquifer.

Previous research regarding oxygen behavior in the vadose zone during wetting and drying cycles (Kim et al., 2004; Dutta
et al., 2015) emphasize the role of the site's operational dynamics in the reclaimed water quality. During the flooding periods
(FP), water content (WC) in the soil profile below the infiltration pond gradually increases (Arye et al., 2011), limiting the
65 scope of diffusive and advective aeration which is crucial for the replacement of the DO used by bacteria. Hence, DP play a
major role in the overall biogeochemical dynamics of SAT systems (Miller et al., 2006). Looking at oxygen dynamics during
wetting and drying cycles in a 1-meter sand column, Dutta et al. (2015) found that DO concentrations dropped exponentially
in response to each flooding event. They also showed that oxygen partial pressure recovered to its initial value upon the start
of the DP. This observation, however, may not hold true for greater depths . In a field scale study, Miller et al. (2006) described
70 oxygen and nitrogen species concentrations in a ~ 2.7 m sandy loam soil profile during cycles of 4 days of wetting and 4 days
of drying. They found that deeper than 0.6 meters below ground surface, aeration was limited compared to the upper parts
of the profile and that deeper than 1.5 meters, the vadose zone was mostly anoxic throughout the wetting and drying cycles
(Miller et al., 2006).

In this study we examine the effect of hydraulic operation (i.e. wetting-drying periods) on a 6-meter soil profile's biogeo-
75 chemical dynamics through a series of long-column experiments. We hypothesize that in vadose zones deeper than ~ 1 -1.5
meters, oxidation-reduction potential (ORP) and DO dynamics differ greatly compared to the shallow parts of a soil profile
and are affected to a different extent by changes in hydraulic management and inflow composition. We further hypothesize
that insufficient DP will have a detrimental effect on the deeper parts of the soil profile and they will eventually result in DO
depletion, negative ORP and impaired outflow quality. Correspondingly, increased DP will be especially significant for the
80 deeper parts of the profile which may lead to extension of the aerobic zone to greater depths. Four main long-column experi-
ments were designed to examine the effect of shorter and longer DP as well as inflow composition on the biogeochemical state
and dynamics of the soil-water system at different depths and the ultimate outflow chemical composition (especially DOC and
nitrogen species).

2 Materials and Methods

85 A 6-meter long, 15 cm diameter, stainless steel column was designed. The column consists of six one-meter modules, each
module is equipped with ports for sensors and sampling equipment. Sensors were located in ports 1, 3, 5, 7, 9 and 13 (Fig.1).
Data acquisition frequency from all sensors was 1 minute. The column was packed with soil from the SHAFDAN site according

to the soil horizons at the site. The different layers' texture as well as initial total organic carbon (TOC) content were determined and are described in the supplementary material (Fahl et al., 2014).



Figure 1. The six-meter long column, Dresden, Germany. Sensors were located in ports 1, 3, 5, 7, 9 and 13

90 Prior to the four main column experiments (Table 1), a preliminary flow experiment was conducted in order to estimate the average flow rate through the column as well as the ponding rate. Average flux during infiltration was estimated to be ~ 0.4 cm/min. The first experiment involved a simple inflow solution of only NH_4^+ (added to tap water, final concentration of ~ 4.5 mg/L of $NH_4^+ - N$), where the hydraulic operation consisted of cycles of 60 minutes of flooding followed by 150 minutes of drying. The term 'flooding period' (FP) refers to the duration of time during which water is pumped to the top of the column. A 'drying period' (DP) starts as the pump is turned off and ends at the beginning of a new flooding event. These specific flooding and drying periods were chosen in light of the preliminary experiment's results, in which it was shown that a DP of 150 minutes is sufficient for the ponding water to infiltrate and allows around 60-70 minutes of free aeration (i.e. no ponding). One of the goals of the preliminary as well as the first experiment was to "awaken" the microbial community of the soil system. The second experiment involved the preparation of a synthetic TWW inflow solution which included NH_4^+ , glucose and asparagine dissolved in tap water. The synthetic TWW composition was designed to include a moderate-to-heavy load of DOC as well as organic and inorganic nitrogen species around the concentrations found in the SHAFDAN ponds. Glucose was chosen as the main carbon source for two reasons: in addition to the fact that it is often used in synthetic WW for laboratory SAT systems (Essandoh et al., 2011; Ak et al., 2013), its high consumption rate by bacteria (compared to more complex carbohydrates or humic material) allowed the investigation of the system's behavior around the ranges of ORP values that are found in field SAT systems (Orgad et al., 2017).

Hydraulic operation for the second experiment included two stages (stages 1 and 2) - we first applied cycles of 60 minutes of flooding and 150 minutes of drying, and after 9 cycles the DP were increased to 240 minutes. During the third and fourth experiments, real TWW water from the Dresden-wastewater treatment plant (WWTP) were used. Hydraulic operation included 60 minutes flooding periods for both experiments and DP were 150 minutes in the third experiment and 240 minutes in the fourth. Experiments 1,2 (stages 1 and 2), 3 and 4 are noted here as AS150, SW150, SW240, RW150 and RW240 respectively, where the abbreviations AS, SW and RW denote the TWW source (Ammonium solution, synthetic TWW and real TWW respectively) and 150/240 denotes the length of the drying periods in minutes.

Table 1. Inflow composition and flooding / drying periods in the four discussed experiments

Experiment	AS150	SW150/240	RW150	RW240
Inflow	NH_4^+ solution	Synthetic WW	Real TWW	Real TWW
FP / DP (min)	60/150	<u>SW150</u> 60/150 <u>SW240</u> 60/240	60/150	60/240

For the synthetic TWW solution the following chemicals were used : L-Asparagine anhydrous ($C_4H_8N_2O_3$, > 99.5%, Sigma Aldrich), Ammonium chloride NH_4Cl (Jenapharm-Laborchemie APOLDA, Analysis pure), D(+)-Glucose monohydrate $C_6H_{12}O_6 \cdot H_2O$ (VWR chemicals PROLABO). The real TWW used for experiments RW150 and RW240 were enriched with glucose and NH_4^+ after initial chemical analysis (presented in the supplementary material) to match the NH_4^+ , TKN and

DOC concentrations to these of the synthetic TWW. Final $NH_4^+ - N$, TKN and DOC concentrations for the synthetic and real WW are resented in Table 2.

Table 2. Chemical composition of the synthetic WW and the real WW after fortification

Analysis	Synthetic TWW	Real TWW
$NH_4^+ - N$	3.8 mg/L	3.8 mg/L
TKN	9.2 mg/L	10.1 mg/L
DOC	103 mg/L	102 mg/L

During all experiments, sensors' information was recorded and pore solution samples were taken from the outflow. Inflow
 120 solution was also sampled and tested to confirm that no major changes in its composition occurred during the experiments. Sen-
 sors along the column included frequency domain reflectometers (FDR) sensors for soil moisture measurement were SM300
 (Delta-T Devices Ltd), submersible Level Sensor (Sensortechincs) was used for Surface head. LDO10101 (Hach-Lange, Ger-
 many) were used for luminescence dissolved oxygen (LDO), and Harburg (ELANA Boden Wasser Monitoring) sensors were
 used to monitor ORP. Tensio 150 (UGT GmbH) tensiometers for pressure head and ECO Tech Bonn (1.5 cm diameter) ceram-
 125 ics were installed along the column as well. While their data is not shown here, it fully supports our presented findings . FDRs,
 WC and DO sensors were located at depths of 25, 75, 175, 275, 375 and 575 cm. ORP sensors were locates in the two upper
 ports (25 and 75 cm depth).

To assess the chemical composition of the inflow solution as well as the collected samples, four types of chemical analysis
 were performed: NH_4^+ was measured using ammonium test kit and a Nova 30 Spectroquant (Indophenol blue method); NO_2^-
 130 and NO_3^- concentrations were measured using an Ion chromatograph (IC) after samples were passed through a 0.2 μm filter;
 TKN was determined using the standard selenium method (ISO 5663:1984) ; Dissolved organic carbon (DOC) was determined
 using the standard method (ISO 8245:1999) after samples were passed through a 0.45 μm filter and HCl was added to prevent
 any further organic matter consumption by bacteria.

3 Results and discussion

135 Figure 2 presents the WC and DO concentrations at four different depths along the profile (25, 75, 175, 275 cm below soil
 surface), during 8 flooding and drying cycles of 60 minutes of flooding and 150 minutes of drying (experiment SW150; WC
 data in a similar manner to Fig.2 for SW240 is presented in the supplementary material). The end of each 60-minutes long
 flooding period is indicated by the 8 peaks of the surface head in each cycle (Fig.2a). It is important to note that the soil surface
 is covered by water for longer than the 60 minutes of flooding - roughly 140 minutes in each cycle, which means the time
 140 where the soil surface is exposed to the atmosphere is roughly 70 minutes. In the second stage of this experiment (i.e SW240),
 where DP were 240 minutes, soil surface was actually exposed to the atmosphere for \sim 160 minutes during each cycle (see
 supplementary material). Water front progression through the profile can be retraced by timing the first WC increase following

the first FP and as reflected by Fig.2, it indicates that flow rate decreased during the first few minutes of the experiment (as should be expected following classic infiltration theory; e.g. Green and Ampt model) but stayed relatively constant as the experiment progressed. WC patterns were significantly different between the various depths - while at a depth of 25 cm WC values ranged between 20.5% and 31.2%, at a depth of 275 cm below the surface, the maximal WC was 18.4% and it dropped below 12% following each DP. Haaken et al. (2016) followed WC patterns in one of the SHAFDAN's infiltration ponds using electrical imaging. While their work is in the field scale, differences in WC between different depths of the profile agree with our findings (Haaken et al., 2016).

DO concentrations also displayed different patterns at different depths of the profile (Fig.2). In the upper parts of the profile (25 cm depth), DO concentration increased in response to each of the DP. In each cycle, the drop in WC caused by the beginning of the DP led to recovery of the DO concentrations and ultimately to complete DO saturation (Fig.2b). As observed in multiple studies in laboratory and field work, close to the surface, DO concentrations are expected to increase in response to the soil aeration during the DP (Mienis et al., 2018; Miller et al., 2006) since regardless of the oxygen movement mechanism (diffusion, advection or convection), the short distance ensures fast response of the system.

At 75 cm depth (Fig.2c), while DO recovery is still observed in response to the beginning of DP, aeration is much less effective and DO concentration averages (through the 8 cycles) dropped by ~ 2.68 mg/L. During the two first cycles, DO concentrations remained almost constant and close to saturation, but upon the beginning of the third DP, a significant decrease was observed. This decrease resulted in a 25% drop in DO concentration following all the next DP. After this initial drop, however, DO patterns and amplitude (following each DP) were sustained through the remaining cycles.

In the deeper parts of the column (Figs.2d and 2e), DP induced DO recovery to a much smaller extent. DO concentrations were able to remain relatively high and constant through the first three cycles, likely due to lower nutrient consumption rates by bacteria at these depths, compared to the upper parts of the profile (Quanrud et al., 1996). Note that DO drop at those depths is a bit hindered, compared to the upper layers. However, as the experiment progressed, DO concentrations decreased steadily to almost complete depletion. These results are to be expected in light of previous laboratory and field observations that demonstrated a decline in DO concentrations with depth (Miller et al., 2006; Orgad et al., 2017). Moreover, DO concentrations along a SAT system's profile are affected by both air movement patterns through the profile during the DP and the consumption of oxygen by bacteria. Since bacterial activity was previously shown to remain relatively unchanged during desiccation (Roberson et al., 1992; Zhang et al., 2012), DO recovery is highly dependent on oxygen transport mechanisms during the DP. These advective and diffusive processes depend on exposure of the surface to the atmosphere, but it also depends on path that air has to pass through, which is longer for greater depths.

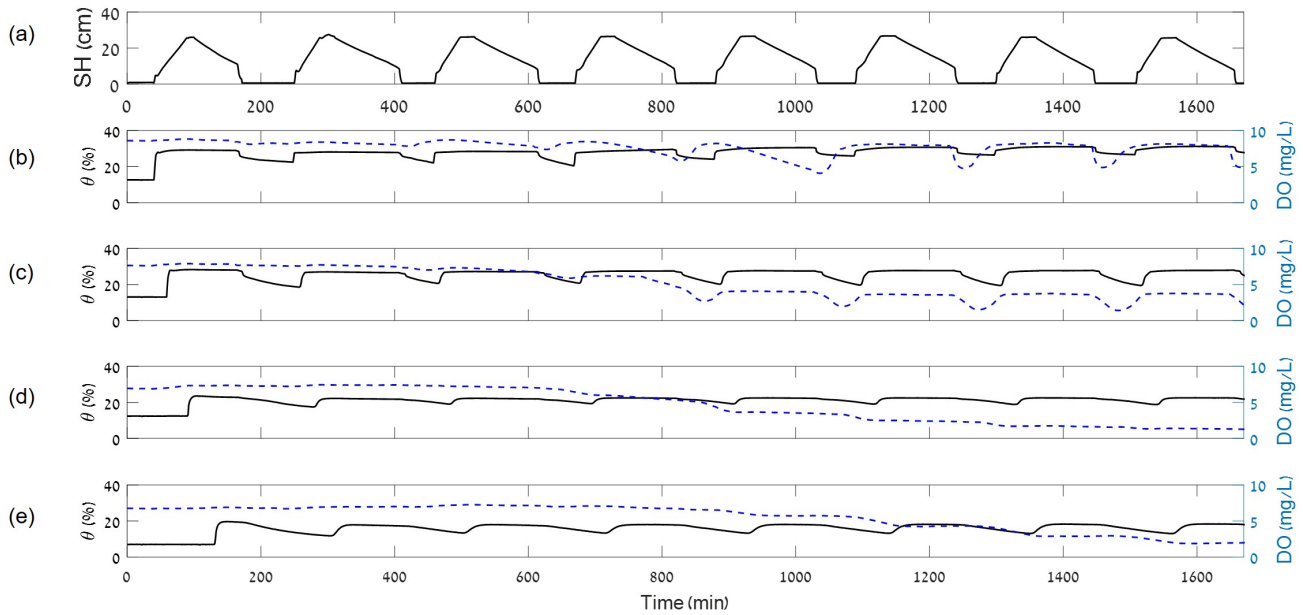


Figure 2. Surface head (SH; 2a), water content (θ) and DO over time at depths of : 25, 75, 175 and 275 cm below soil surface (2b-2e respectively) during SW150

As reflected by the results of SW150 (Fig.2), the 150-minute DP resulted in significant DO depletion in depths greater than 75 cm. To examine the effect of a change in hydraulic operation (longer DP), we increased the length of the DP to 240 minutes (60% increase) after the 10th cycle. Figure 3 presents the DO concentrations at four different depths in the soil profile through the two-stage flooding-drying campaign (experiments SW150 and SW240). By increasing the DP, we increased the time of soil surface's exposure to the atmosphere to ~ 160 minutes on average (compared to ~ 70 minutes on average for SW150) and thus expected to observe increased aeration and some recovery of DO concentrations deeper than 75 cm (where the shorter DP led to almost full DO depletion). During SW240, WC values at a depth of 75 cm below the surface ranged between $\sim 18\%$ and $\sim 12\%$ (compared to $\sim 27\%$ and $\sim 17\%$ for SW150). At depths of 175 and 275 cm below ground surface, maximal WC were ~ 22 and 18% , respectively, and minimal WC were ~ 16 and 11% respectively. At 75 cm depth, shortly following the first longer DP (~ 100 minutes), DO concentrations increased by $\sim 60\%$ and reached an average of ~ 6.4 mg/L following each of the next DP (compared to an average of ~ 3.7 mg/L during the shorter DP, after the initial drop around the third cycle). In the deeper parts of the profile, a delayed, moderate yet significant response to the increase in DP was observed. The delay in response time compared to the DO recovery in the upper most part of the profile, increased with depth, corresponding to the expected dynamics of air and oxygen movement through the soil profile (DO recovery was observed after around 2,700 minutes for 375 cm and 3,700 minutes for 575 cm). The minimum DO concentrations recorded through the two-stage experiment averaged 0.79, 1.32 and 1.62 mg/L for depths of 175, 375 and 575 cm, respectively. These ranges of DO concentrations below 1.5 meters underground are found in many SAT sites around the world (Amy et al., 2007) and explain the negligible aerobic

bacteria activity found under these conditions.

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Following each of the longer DP, DO concentrations in the 175 cm sensor fluctuated periodically in response to the wetting and drying periods and reached 4.1 mg/L following the drying events. In the two deepest sensors, DO concentrations stabilized ~ 3.9 mg/L in the 375 cm sensor and ~ 3.25 mg/L in the 575 cm sensor. Considering the fact that sustaining the shorter DP of SW150 would result in total DO depletion ~ 175 cm depth (supplementary material), these are very important observations.

195 Clearly, too short DP will lead to reducing conditions, at some depth, which as a consequence may lead to insufficient degradation of residual DOC, nitrogen species, and possible leaching of undesired minerals and compounds, such as manganese to the deep vadose zone and the aquifer (Goren et al., 2012). In addition, the prominent increase in DO in the deeper parts of the profile indicate that longer DP means not only enhanced oxidizing conditions at a given depth, but also increase of the profile volume that did not develop anoxic or anaerobic conditions over the flooding and drying cycles.

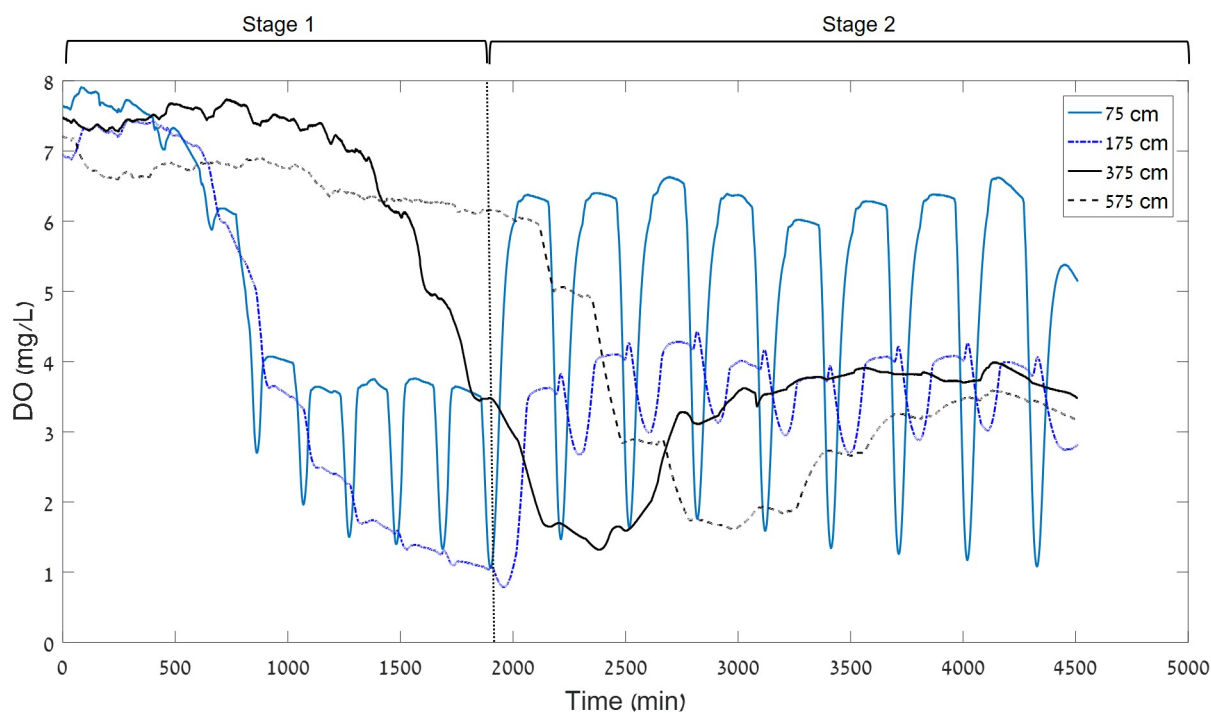


Figure 3. DO concentrations at depths of 75, 175, 275 and 575 cm in response to the increase in DP (between SW150 and SW240). Note the convergence of the deep sensors to < 2 mg/l during the short DP versus convergence to > 3 mg/l in response to the longer DP.

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Experiments RW150 and RW240 were designed to further examine the effect of the difference in DP duration on the biogeochemical state of the profile and its effect on some water quality parameters. In these experiments we used real TWW, collected from the Dresden wastewater treatment plant (WWTP) after the activated sludge process. Similarly to what we observed in SW150 and SW240, longer drying periods had an advantage in terms of DO concentrations along the profile

(not shown). ORP measurements in the upper parts of the column (75 cm) revealed that ORP was significantly higher during RW240, ranging between $\sim +400$ and $\sim +160$ mV compared to RW150, during which ORP was mostly negative and reached values as low as ~ -530 mV (supplementary material).

Figure 4 presents $NH_4^+ - N$, $NO_3^- - N$, DOC and TKN and concentrations at the outflow (i.e. 6 meters below soil surface), comparing the results for RW150 and RW240. Outflow $NH_4^+ - N$, TKN and DOC concentrations during RW240 (~ 0.033 , ~ 0.62 and ~ 1.65 mg/L respectively) were significantly lower compared to their inflow concentrations. During RW150, $NH_4^+ - N$, TKN and DOC outflow concentrations (~ 0.5 , ~ 3.8 and ~ 4.4 mg/L, respectively) were also lower compared to the inflow, but averaged significantly higher compared to RW240 (t-test, $\alpha=0.05$), suggesting that the longer DP had a significant positive effect on the outflow quality.

During RW150, $NH_4^+ - N$ concentrations were ~ 0.1 mg/L after ~ 400 minutes from the beginning of the experiment but increased noticeably in the following samples (taken after ~ 1500 minutes) while $NO_3^- - N$ concentrations decreased correspondingly. Considering the DO concentrations recorded during the experiment, this observation is to be expected. Progressively decreasing DO levels were observed at all depths greater than 75 cm starting after ~ 850 minutes from the beginning of the experiment (see DO data for depths of 175 and 275 cm in the supplementary material). This suggests that the DO depletion caused by restricted aeration for this duration of time, led to decreased rates of nitrogen species' oxidation, which may explain the increase in $NH_4^+ - N$ concentrations at the outflow after ~ 1500 minutes. However, looking at the TKN analysis for this experiment, we found that TKN did not significantly increase, suggesting that the increase in $NH_4^+ - N$ concentrations occurred simultaneously to a decrease in organic nitrogen concentrations. This behavior may theoretically be attributed to increased rates of ammonification. However, ammonifying bacteria populations have been shown to thrive under DO saturation (Ruan et al., 2009), which makes this possibility unlikely. Since this study did not include microbial identification, further investigation is needed in order to reveal the exact nature of this observation.

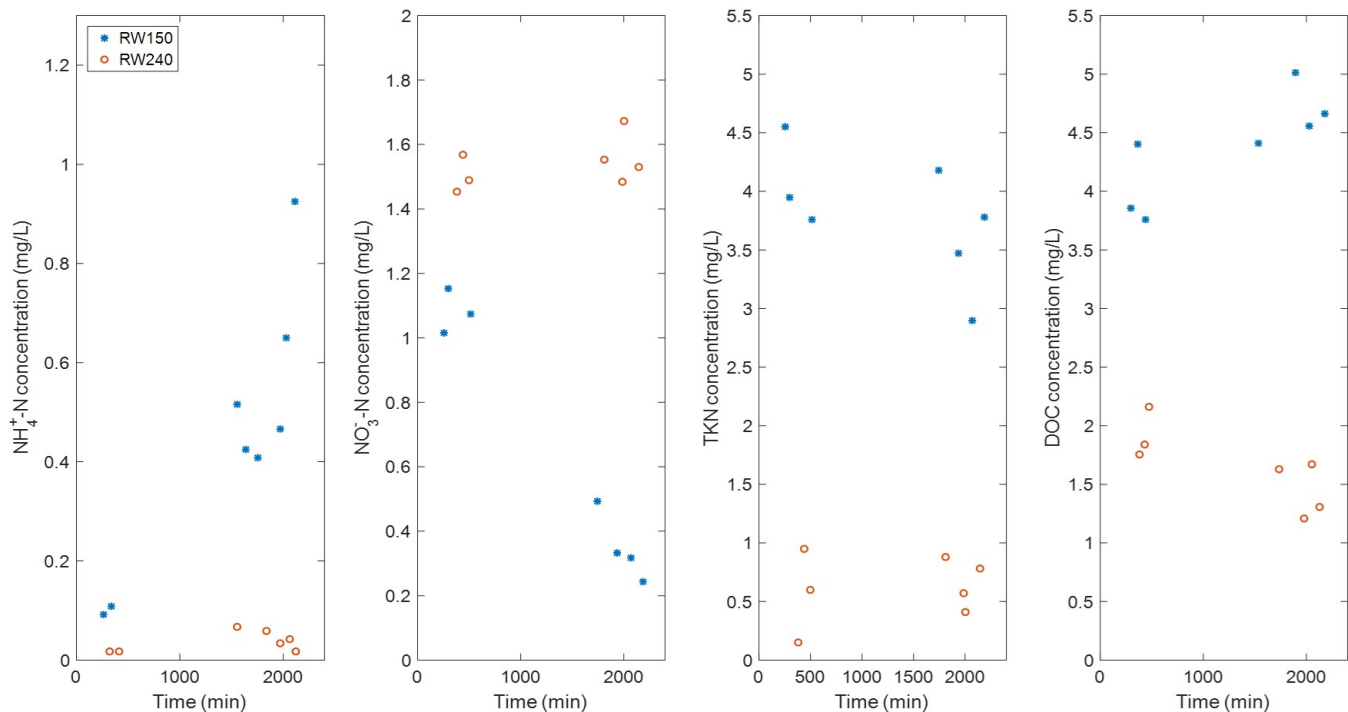


Figure 4. Outflow concentrations of $NH_4^+ - N$, $NO_3^- - N$, TKN and DOC (mg/L) , during shorter (blue) and longer (red) DP (experiments RW150 and RW240 respectively)

3.1 Comparison with field observations

In a field study conducted in the Israeli SHAFDAN site in 2015-2016, Orgad et al. (2017) recorded ORP values along the wetting and drying cycles in a series of flooding and drying campaigns over a year. A description of the site operation and characteristics is brought by Orgad (2017), Idelovitch et al. (2003), Icekson et al. (2011) and others. Figure 5a presents the maximal ORP values in these campaigns at a depth of 95 cm below the pond's surface, around 20 m south-east of the inlet. The ensemble of wetting-drying cycles over an entire year assures a wide variety of wetting and drying proportions. The recorded data showed that when DP were ~ 15 hours or longer, the maximal ORP values consistently indicated aerobic conditions (and reached $\sim + 650$ mV).

Figure 5b presents the same relationship for the column experiments presented above. Despite the major differences in time scales (order of 2-4 hours for column experiments vs. order of 15 hours for field conditions), TWW composition as well as system's structure and characteristics between our 1D system and a 3D full scale SAT site, our measurements (WC, DO, ORP) qualitatively agree with the field observations. That is, longer DP led to higher (oxidizing) ORP values, while shorter DP led to lower ORP values (reducing conditions). Comparing the ORP values between the field and the column results (Figs.5a and 5b), it is clear that there is a non-negligible difference - while ORP values never exceeded +440 mV in the column experiment,

values of $\sim +700$ mV were observed in the field. Moreover, minimal ORP values did not drop below $\sim +100$ mV in the field while in the column, negative values of ~ -150 mV were observed. These differences probably stem from the fact that in the column, a 1D system, air (and water) flow only along the vertical direction while in the field, air flows through the soil profile in all directions and thus allow overall enhanced aeration. Further, in the field the infiltration pond is filled from a single inlet, which means it takes about 10 hours for full surface coverage of the pond. This suggests that further from the inlet, lateral air-flow is possible even hours after the initial flooding.

The difference in time scales between the column and the field experiments may also suggest a difference in the proportions of the air supply mechanisms to different layers of the subsurface. Mizrahi et al. (2016) have suggested that air (in the gas phase) may be "pushed" downwards once the soil surface is fully flooded by water. While this phenomenon is likely maximized in a column experiment (as the soil-air has no time and no horizontal pathway to be released), this mechanism and its impact were not investigated here. However, it is likely that in field conditions, where lateral air movement is allowed, this mechanism of air supply is of lesser significance, and air movement due to diffusion or air-pressure gradients becomes more dominant.

Interestingly, it is evident from the field data (Fig.5a) that very long DP (>20 hours) did not present an advantage over DP of moderate duration (between 15 and 20 hours), at least at a depth of 95 cm. Considering the common understanding that most of the biochemical degradation of organic matter at the SHAFDAN SAT site happens at the upper meter or so (Quanrud et al., 1996), DP of more than 15-20 hours seem to have no influence on the oxidation state of the profile. In light of the understanding that longer DP lead not only to increased aeration but also to an increase of the aerobic volume of the soil profile, a combination of longer FP and sufficiently long DP (such that allow this profile aerobic volume increase) may be beneficial for both amounts of TWW that the site is able to treat per unit time as well as reclaimed water quality.

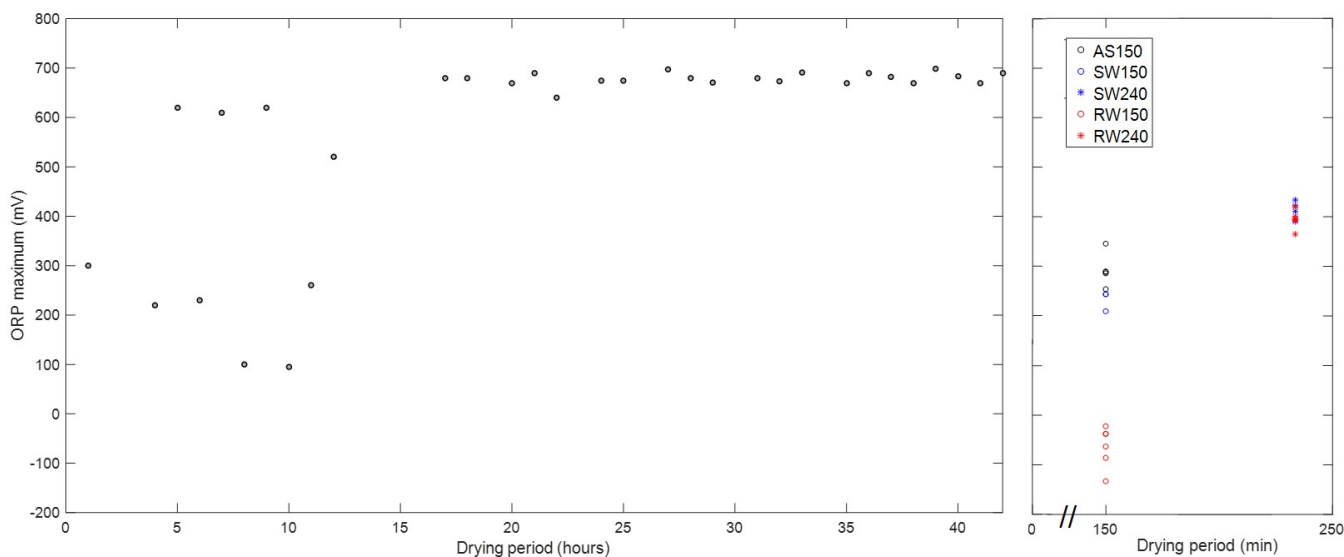


Figure 5. Maximal OPR values versus DP length a) during field infiltration campaigns in the SHAFDAN site (at 95 cm depth) and b) during the column experiments AS, SW150/240, RW150 and RW240

260 As described in Table 1, two of the four experiments presented in this work (RW150 and RW240) included real TWW while in the two earlier experiments synthetic TWW were used. To allow comparison between the experiments, their inflow DOC, TKN and $NH_4^+ - N$ concentrations were matched (see supplementary material) so that the main difference between the two WW sources stems almost entirely from their microbial content (likely there are some differences in micro-nutrients as well). Compared to tap water-based inflow that contained very low concentrations of bacteria, the real TWW collected
265 from the Dresden WWTP had gone through an activated sludge reactor but were not disinfected. Hence, it contained much larger quantities of microorganisms. During RW150, we observed lower ORP values compared to SW150 that is identical in terms of FP/DP. The average peak ORP throughout the SW150 were $\sim +220$ mV while in RW150, the average peak ORP was ~ -80 mV (Fig.5b), despite the fact that the inflow in both experiments contained similar organic loads. Since both RW150 and SW150 had similar DPs, the lower overall ORP values recorded in RW150 suggest that the difference in ORP stems from
270 oxygen consumption rates. Theoretically, difference in oxygen consumption rates may stem from either higher concentrations of bacteria or the presence of less easily degradable organic carbon sources in the WW (compared to glucose). Ak et al. (2013) compared organic carbon degradation and DO concentrations between synthetic and real wastewater infiltration in a series of 120 cm soil column experiments. They found that degradation of organic carbon was more efficient for the synthetic WW which they attributed to the presence of glucose as the main carbon source in the synthetic WW (compared to the less
275 easily biodegradable carbon sources in the real WW). In our experiments, however, the real TWW collected from the Dresden treatment plant had only ~ 10 mg/L of organic carbon prior to the addition of glucose (i.e. glucose accounted for $>90\%$ of the DOC in the experiments with real TWW). This may suggest that the lower ORP values recorded in RW150 are related to additional oxygen consumption by the microbial community in the influent itself which therefore may be more dominant in the degradation process than commonly perceived.

280 In field scale SAT systems, clogging is an important issue to consider. Throughout the long-column experiments described here, we did not observe significant indications of physical or biological clogging. This is evident by the consistent patterns of surface head along the flooding and drying cycles. Possibly, this is due to the fact that the experimental setup was not exposed to sunlight and therefore algae development is not expected. However, the development of biofilm has an effect on the hydraulic properties of the unsaturated zone (Volk et al., 2016) and since it has been shown that the organic load of the influent
285 is positively correlated to the phenomenon of bioclogging (Rosenzweig et al., 2014), this will be an important topic to address when dealing with full scale sites.

4 Summary and conclusions

In a series of long column experiments we examined an SAT system response to different hydraulic operation regimes, namely wetting and drying periods. Experiments were conducted using real and synthetic treated wastewater. WC, DO and ORP were
290 monitored along the column, water samples were collected at the outlet, and analysed for nitrogen species and DOC. Hydraulic regimes considered were of 60 minutes of wetting, and 150 or 240 minutes of drying (corresponding to $\sim 140/70$ and $\sim 140/160$ minutes of the soil surface being ponded/exposed to the atmosphere, respectively).

As could be expected, shorter drainage periods led to higher WC regimes in the subsurface, and lower DO values. Interestingly, the shorter DP also led to almost complete depletion of the DO throughout the column (below ~50 cm), and creation of prevailing low ORP values, indication anoxic to reducing conditions throughout most of the column. Longer DP, on the other hand, led to oxidizing conditions throughout the column for most of the time. These conditions are also expressed as column outlet water qualities, where longer DP led to better oxidation of NH_4^+ , and reduction of total nitrogen as well as DOC. The almost immediate response of the DO concentrations at depth to the surface exposure time to the atmosphere suggests domination of advective oxygen movement in the gaseous phase (over diffusion in the gaseous phase or advection in the liquid phase). These results suggest that longer DP not only shift the active (oxidizing) part of the system towards oxidizing conditions, but it also makes this region larger. To the best of our knowledge, this is the first work to suggest dependence of the aerated part of the vadoze zone under SAT upon the DP, rather than a simple consideration of higher aeration of a fixed volume close to the surface.

Longer DP have their consequences - on one hand, they led to more oxidizing conditions, and to the extension of the oxidizing parts of the variably saturated zone. On the other hand, they led to reduction in the volumes that can be infiltrated and recharged to the aquifer. Clearly these are contrasting objectives that need to be further explored and optimized for a given site. The qualitative similarity between ORP values in these column experiments and those measured at the SHAFDAN site suggest that these findings are also relevant for real conditions, and that such optimization can be conducted also in reality.

To conclude, our results suggest that the variably saturated zone in a SAT system can and should be seen as a pseudo reactor, in which DO and ORP values can, to a certain extent, be controlled. Longer DP allowed better degradation of NH_4^+ , organic nitrogen and organic carbon, but led to reduced infiltration. The immediacy of the DO recovery at depth following sufficient DP may suggest that the ratio between drying and wetting periods is not the parameter that dictates the SAT biogeochemical dynamics, and long enough DP (that is primarily a function of hydraulic properties and desired oxidizing depth) can be followed by much longer wetting periods. This however was not examined, yet.

Code and data availability. The data that support the findings of this study are available from the corresponding author, SBM, upon reasonable request.

Author contributions. FB and JS prepared the experimental set up, SBM, AF and NW designed the experiments, OO provided and analysed the field data, SBM performed the column experiments, analysed the data and prepared the paper with contribution of all authors.

Competing interests. The authors declare that they have no conflict of interests

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