- 1 Continuous Monitoring of a Soil Aquifer Treatment System's Physico-Chemical
- 2 Conditions to Optimize Operational Performance

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# Highlights

- Long wetting stages reduce soil percolation capabilities during winter.
- Redox and gaseous O<sub>2</sub> display intensive dynamics in the top 25 cm of the soil aquifer treatment vadose zone.
- Optimal wetting and drying stages are defined according to E<sub>h</sub> and gaseous O<sub>2</sub>
  observations.
- The length of wetting and drying stages should be defined separately rather than by adhering to their ratio.

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### **Abstract**

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Soil aquifer treatment (SAT) is a tertiary process for wastewater treatment where the wastewater 25 infiltrates through a thick vadose zone for purification and storage in the underneath aquifer. 26 27 SAT infiltration basins are typically flooded intermittently, while maintaining a fixed ratio 28 between the wetting and the drying stages. However, infiltration basins exhibit different physical and chemical properties, limiting the generalization of SAT operation to attain optimal 29 30 efficiency. Since frequent sampling of the soil pore water to verify the SAT's biodegradation efficiency can be arduous, continuous monitoring of the SAT vadose zone's physico-chemical 31 32 conditions is required. In this study, redox potential (E<sub>h</sub>) was continuously monitored, together 33 with other variables such as volumetric water content  $(\theta)$ , soil temperature, and gaseous oxygen (O<sub>2</sub>), at multiple depths of a SAT vadose zone throughout the year and while the system was 34 35 constrained to different operational modes. Hydrological models were calibrated and validated to water content observations, and they illustrated the seasonal changes in water infiltration. 36 37 Furthermore, it was shown that under long wetting stages during winter, there was a reduction in the SAT's drainage capabilities. The E<sub>h</sub> observations, under long wetting stages, demonstrated 38 39 larger variability and very negative values as ambient temperature increased. Assembling the daily E<sub>h</sub> observations illustrated that a wetting stage should cease after about 30 hours, once 40 41 suboxic conditions are established. A drying stage's optimal duration should be 36 hours, according to the E<sub>h</sub> and O<sub>2</sub> observations during summer and winter. Ultimately, the study shows 42 that the length of wetting and drying stages should be defined separately, rather than by adhering 43 to the wetting/drying ratio. 44 45

#### 1 Introduction

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Worldwide water scarcity has motivated the development of alternative water resources such as 48 the reuse of treated wastewater. Soil aguifer treatment (SAT) is commonly implemented to 49 50 further improve the recovered water's quality and remove the majority of suspended matter, 51 microorganisms, viruses, and organic and inorganic constituents (Dillon, 2005; Goren et al., 2014; Massmann et al., 2006; Schmidt et al., 2011; Tsangaratos et al., 2017). In SAT systems, 52 the treated wastewater is recharged to the underlying aquifer by surface spreading over 53 54 infiltration basins. The wastewater is purified mainly through the physical and biochemical 55 processes that occur during water passage through the vadose zone (Dillon, 2005; Elkayam et al., 56 2015). Although SAT systems have been used for decades (Grinshpan et al., 2021; Bouwer, 2002), the ability to estimate and predict a SAT system's performance is still challenging, and 57 58 the optimal SAT operation is still under investigation (Ben Moshe et al., 2020; Sharma and Kennedy, 2017). 59 A major uncertainty in SAT systems concerns the vadose zone processes that play a central role 60 in determining the quality of the water that recharges the aquifer (Elkayam et al., 2015). The 61 chemistry of the percolating wastewater changes due to a combination of several biogeochemical 62 processes, such as organic matter biodegradation, nitrification, sorption, cation exchange, etc. 63 (Amy and Drewes, 2007; Díaz-Cruz and Barceló, 2008; Goren et al., 2014; Miller et al., 2006; 64 Tufenkji et al., 2003). Most of the organic matter is removed by biodegradation (i.e. microbial 65 activity) within the upper two meters of the vadose zone (Drewes, 2009). Nevertheless, the 66 microbial activity is greatly affected by the soil water content, which frequently changes in SAT. 67 68 Generally, a major challenge in SAT systems is to facilitate the intrusion of  $O_2$ , primarily in the 69 gaseous phase, and to enrich the active subsurface with O<sub>2</sub> (Ben Moshe et al., 2020; Massmann et al., 2006). 70 A consequence of the perturbation in the O<sub>2</sub> supply to SAT is expressed in changes in the redox 71 conditions (Mächler et al., 2013; Rezanezhad et al., 2014). Redox potential or oxidation-72 reduction potential is a quantitative measure of electron availability, i.e., the tendency of the 73 system to receive or donate electrons (Hinchey and Schaffner, 2005). Substantial changes in 74 SAT systems' redox conditions might lead to the release of undesirable metals, such Fe<sup>2+</sup> and 75

Mn<sup>2+</sup> (Goren et al., 2012), and affect the degradation rates of pesticides and pharmaceutical 76 77 substances (Massmann et al., 2006). Additionally, previous studies have illustrated the possible 78 degradation of groundwater quality due to the emergence of contaminants that leach from the SAT vadose zone under reducing conditions (Asano and Cotruvo, 2004; Massmann et al., 2006; 79 Oren et al., 2007; Sharma and Kennedy, 2017). Redox processes are associated with the 80 degradation of organic matter by terminal electron acceptors or redox couples, such as O<sub>2</sub>/H<sub>2</sub>O, 81 NO<sub>3</sub>/N<sub>2</sub>, MnO<sub>2</sub>/Mn<sup>2+</sup>, Fe<sup>3+</sup>/Fe<sup>2+</sup>, and SO<sub>4</sub>/H<sub>2</sub>S, in sequential order from the highest energy yield 82 downwards (Berner, 1981; Froelich et al., 1979; Christensen et al., 2000). The transition between 83 84 redox conditions is determined by the presence and availability of these electron acceptors/donors. Once the strongest oxidizing species (O<sub>2</sub>) is depleted, the next strongest 85 oxidizing species is used (NO<sub>3</sub>) and so on. The alternation between oxic (> 400 mV), suboxic 86 87 (between 400 and -100 mV), and anoxic (-100 mV >) conditions in the vadose zone depends on the availability of the oxidized species (Reedy et al., 2000). In addition, studies have reported on 88 89 the seasonal (temperature changes) effects on redox conditions, which were attributed to the increase in dissolved oxygen concentrations at low temperatures (Massmann et al., 2006) and the 90 91 greater microbial activity (i.e., higher O<sub>2</sub> consumption) at higher temperatures (Greskowiak et al., 2006; Kirschbaum, 1995). 92 An important operational aspect of a SAT system is the intermittent application of the effluents 93 (Sattar, 2016; Sallwey et al., 2020). After the infiltration basin is flooded with wastewater, a 94 95 drying period is implemented to sustain the SAT's infiltration capacity and biochemical 96 capabilities (Sharma and Kennedy, 2017). The wetting and drying stages, which can be expressed by the wet/dry ratio parameter, have a critical impact on the removal rates of dissolved 97 organic carbon, total nitrogen, and pathogens (Ben Moshe et al., 2020; Morrison et al., 2020; 98 Sharma and Kennedy, 2017). Although the wet/dry ratio can vary depending on location and 99 wastewater quality, it is well accepted that it should be below 1.0 (Sattar, 2016; Sharma and 100 Kennedy, 2017). Nevertheless, infiltration basins behave differently with regard to infiltration 101 rates and clogging. Thus, in many cases, the SAT operational efficiency is limited to the personal 102 103 experience of the operators and their knowledge of the specific infiltration basin (Sharma and Kennedy, 2017). Note, however, that several studies (e.g., Ben Moshe et al., 2021b) suggest that 104 it is not the wet/dry ratio that should be considered, but specific times for wetting and drying. 105

The oxidation-reduction potential (Eh), together with chemical and physical parameters such as water content, soil temperature, O<sub>2</sub> concentration, etc., can be continuously monitored by installing the relevant sensors. Previous studies have implemented the Eh sensor and successfully described, with high temporal resolution, the subsurface chemical conditions in various environments, such as wetlands, the groundwater (or the capillary fringe), aquifers, etc. (Wallace et al., 2019; McMahon and Chapelle, 2008; Shenker et al., 2005; Silver et al., 2018; Rezanezhad et al., 2014). To improve SAT system performances, the link between the wetting and drying stages and the subsequent redox conditions developed in the subsurface should be established. Thus, in situ monitoring can improve SAT management performance and reduce the subjectivity of the operator. The objective of this study was to examine the temporal variability in redox potential and the way it is affected by changes in volumetric water content, gaseous O<sub>2</sub>, and climate imposed by different operational modes of wetting and drying stages. Furthermore, calibrated and validated hydrological models were used to explore the behavior of water fluxes under different operational modes and seasonal temperature changes. Finally, the optimal lengths of a drying stage and a wetting stage were determined, following the in situ observations.

### 2 Methods

## Study sites

The Dan region reclamation project (Shafdan) reclaims about 125 million m³ of effluent annually, from the Tel Aviv metropolitan area in Israel. The treatment of effluents occurs in two stages. The first stage involves mechanical-biological treatment, which is based on activated sludge, while in the second stage, the treated water (a secondary effluent) is delivered to infiltration basins, as part of the SAT system, to further improve water quality. Six infiltration basin sites, covering a total area of 1.053 km², are located in central Israel, overlying the coastal aquifer (Figure 1). Each basin is divided into several spreading ponds, about 1500 m² each, which are alternately flooded. The vadose zone that underlies the basins is mostly composed of sand, sandy loam soil, and calcareous sandstone layers. Typically, the ponds are flooded for one to two days (max hydraulic head of about 50 cm), followed by two to six days of drainage and soil surface drying. The wetting and drying stages are controlled by the ponds' flooding order,

the availability of effluent, and the drying period, which is suggested to be at least 24 h (Icekson-

Tal et al., 2003). The basin surface is plowed on a regular basis to break up the developed

biocrust and to prevent clogging (see Negev et al. (2020) for details).

# Study operation

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139 In this research, two in situ measurement stations were installed in an infiltration pond during 2018 (pond 4103 in the Yavne 1 cluster, Figure 1). Each station was equipped with several sets 140 of sensors at 25, 50, 75, 100 and 150 cm depth, including time domain transmittance (TDT) 141 probes (Acclima Inc., Idaho, USA), copper-constantan thermocouples (OMEGA Engineering, 142 Inc., Connecticut, USA), oxidation-reduction potential (ORP) electrodes (ELH016 van London 143 Co, Houston, TX, USA), and O<sub>2</sub> percentage probes (ICT02 sensor, ICT Int., Australia). Data 144 were collected at prescribed intervals and logged on a CR1000 data logger (Campbell Scientific, 145 Inc., Logan, UT, USA). In addition, suction cups were installed at similar depths. In station 1 146 147 (Figure 1), the data consisted of volumetric water content ( $\theta$ ), soil temperature (T), and ORP (E<sub>h</sub>) time series, which were continuously measured every 20 minutes between 28/07/2020 and 148 10/02/2021 (total of 14,185 values, 197 days, for each variable). The data were obtained at 25, 149 50, and 100 cm depths. In station 2 (Figure 1),  $\theta$ , T, gaseous oxygen (O<sub>2</sub>), and E<sub>h</sub> were measured 150 every 20 minutes between 08/05/2019 and 20/07/2020. There were about 60 days in which data 151 were not collected in station 2 due to technical issues. The data from station 2 contained 27,222 152 points of  $\theta$ , 29,394 points of  $O_2$ , 30,414 points of  $E_h$ , and 26,730 points of T measurements. In 153 station 2, the data were collected at 25, 50, 75 and 100 cm depths. 154 The water quality characteristics of the secondary effluent that flooded the Yavne 1 basin are 155 presented in the Supporting Information (Fig. S1). Note that the quality parameter concentrations 156 conform to the updated "Inbar" regulations (Inbar, 2007) and the findings of a previous study 157 that surveyed numerous wastewater storage and treatment reservoirs across the country (Kfir et 158 al., 2012). To determine the soil physical properties, undisturbed soil cores were sampled at 159 different depths. Subsequently, flow experiments were carried out to calculate the saturated 160 161 hydraulic conductivity (Ks) based on Darcy's Law (Supporting Information, Fig. S2). Additionally, particle size distribution (PSD) analyses are presented in Fig. S3 (Supporting 162 *Information*). The PSD results indicate that the SAT vadose zone is homogeneous. 163

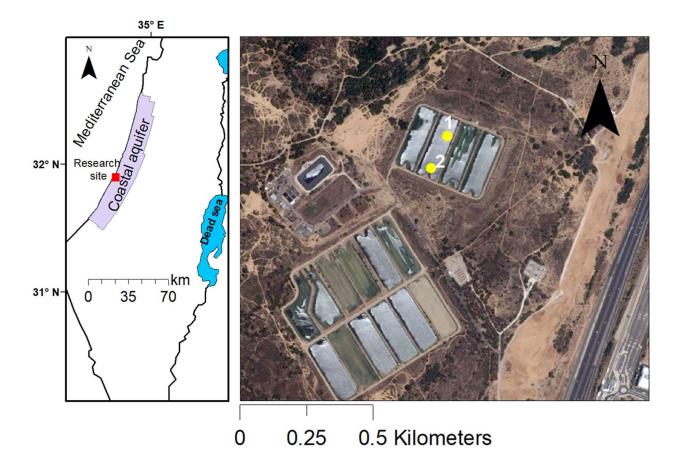


Figure 1: The location of the investigated site, the Yavne 1 infiltration basin of the Shafdan. In the close up of the investigated pond, the yellow circles represent the locations of the measurement stations (©Google Earth).

# Hydrological model and gaseous oxygen dynamics

The calculations for water and oxygen fluxes in the SAT vadose zone are calculated differently for the ponding stage and for the stage where there is no ponding (water) on the soil surface. For the ponding stage, Ganot et al. (2017) showed that the infiltration rates in managed aquifer recharge systems can be predicted reasonably well by simple analytical models. In this study, the Green-and-Ampt equation for infiltration into a flooded soil was implemented to calculate the water flux (Bouwer, 2002):

$$q = Ks \times \frac{(L + d - \psi^*)}{L} \quad (1)$$

where q (L T<sup>-1</sup>) is the water flux, d (L) is the ponding depth, L (L) is the thickness of the saturated vadose zone, and  $\psi^*$  (L) is the negative pressure head at the wetting front. Note that L is assumed here to be constant, the subsurface is assumed to be homogeneous, and  $\theta$  is assumed to vary with time only. As the wetting front progresses, the gradient approaches a value of unity, and the infiltration rate becomes equal to the hydraulic conductivity of the wetted zone. Once the water ponding ceases, the water drainage is set equal to the unsaturated hydraulic conductivity, described with an exponential form (Guswa et al., 2002):

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$$D(\theta) = Ks \frac{e^{\beta(\theta - \theta_{fc})} - 1}{e^{\beta(\theta_s - \theta_{fc})} - 1} \quad (2)$$

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where Ks (L T<sup>-1</sup>) is the saturated hydraulic conductivity,  $\beta$  is a parameter of the soil,  $\theta$  (L<sup>3</sup> L<sup>-3</sup>) is the volumetric water content,  $\theta_{fc}$  (L<sup>3</sup> L<sup>-3</sup>) is the water content at field capacity, and  $\theta_s$  (L<sup>3</sup> L<sup>-3</sup>) is the saturated water content. Furthermore, the effect of temperature changes on the soil hydraulic conductivity is implemented through the change in viscosity (Lin et al., 2003):

$$Ks_T = Ks_{25} \frac{\mu_{25}}{\mu_T}$$
 (3)

- where  $Ks_T$  and  $Ks_{25}$  are soil hydraulic conductivity values at temperature T°C and 25°C, respectively, and  $\mu_T$  and  $\mu_{25}$  are the dynamic viscosity of water (M L<sup>-1</sup> T<sup>-1</sup>) values at T°C and 25° C, respectively.
- An inverse problem was set to find an optimal combination of Ks and  $\beta$  parameters that minimizes the following objective function:

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$$\Phi(b) = \sum_{i=1}^{N} [\theta(t_i) - \theta(t_i, b)]^2 \quad (4)$$

where N is the number of the  $\theta$  observations,  $\theta(t_i)$  are the observations at a specific time, and  $\theta(t_i,b)$  are the corresponding models' (Eq. 1 to 3) predictions for the vector of optimized parameters, b (Ks and  $\beta$ ). The inverse problem was solved using the *fminsearch* function in MATLAB. To evaluate the prediction quality, the root mean squared error (RMSE), the Nash

Sutcliffe efficiency (NSE), and the Pearson correlation (r) were calculated following Ritter & Muñoz-Carpena (2013).

#### 3 Results and Discussion

The water level measurements at the soil surface were collected by the Shafdan operators as part of the operational routine. Analysis of the water level data (ponding depth) indicates two operational modes of long and short cycles that were implemented at the investigated pond. The characteristics of the drying and wetting stages, as recorded by the operators, are summarized in Table 1. Note that the wetting stage is defined when water is present at the soil surface and the drying stage is defined when water is absent. Further note that the water level is measured at a single point (close to the pond inlet). Therefore, some variations, at the order of a few cm, may exist due to microtopography and the distance between the inlet and the far parts of the pond. This may lead to some delays in water arrival or recession from the stations. Throughout the analysis described below, we define the winter period as the months between November and April and the summer period as the months between May and October, corresponding to the Mediterranean climate. Our monitoring systems are operated independently from the Shafdan facilities.

**Table 1.** Technical information of the recorded long and short wetting and drying cycles (this data was provided by the Shafdan operators).

	Long cycles	Short cycles
Wetting stage (days)	9 ± 2.4	$1.5 \pm 0.4$
Drying stage (days)	$3.3\pm2.4$	$1.8 \pm 1$
Number of recorded cycles	33	37
Length of cycle (days)	$12.7\pm3$	$3.2\pm1.3$
Wet/dry ratio	$3 \pm 1.8$	$0.9 \pm 0.3$

# Hydrological conditions

A representative set of  $\theta$  time series measured in the SAT's vadose zone is presented to describe 222 the variability in hydrological conditions measured throughout different seasons and operational 223 224 modes (Figure 2). The  $\theta$  measurements were obtained during long (Figure 2a, b, c) and short (Figure 2d) cycles at three different depths. Note that the water content measurements presented 225 in Figure 2 (a, b) were recorded at station 2 (Figure 1), during summer (Figure 2a) and winter 226 227 (Figure 2b). The water content variations under short cycles were measured at station 1 (Figure 2c, d). Differences in the absolute values between the water content observations at different 228 depths are mainly related to the vertical texture variability (Fig. S2, Supporting Information). 229 Under the long cycles,  $\theta$  measurements were obtained throughout 19 cycles during summer 230 (May-October) and 14 cycles during winter (November-April). Under short cycles, there were 231 12 cycles during summer and 25 during winter. 232 233 Every recorded wetting event prompted an intensive infiltration that was expressed by a rapid and almost instantaneous increase in water content at all depths and under the two operational 234 235 modes (Figure 2). Furthermore, the soil remained at similar level of saturation throughout each wetting stage. Similarly, once the drying process started, it occurred virtually simultaneously at 236 237 all depths. There were noticeable differences in the drainage rates between summer and winter, where the soil dried faster in summer. To elaborate the drainage process, the drying stages were 238 239 assembled and averaged at an hourly interval and separated into short (Figure 3a) and long (Figure 3b) cycles. Additionally, Eq. (1), (2), and (3) (the hydrological model) were implemented 240 to describe the water flow in the SAT's vadose zone under long and short cycles. The 241 hydrological models were calibrated and validated against water content observations at 25 cm 242 243 depth by adjusting the Ks and  $\beta$  parameters (Fig. S4 and Fig. S5, Supporting Information, Table 2). Throughout the calibration, the Ks and  $\beta$  parameters attained different values for the long 244 cycle periods and the short cycle periods (Table 2). There are differences in soil physical 245 properties between stations 1 and 2, which explains the need for calibrating different parameter 246 sets. In addition, the calibrated Ks values for both models were substantially lower than the 247 measured Ks values (Fig. S2, Supporting Information, Table 2). It has been shown that Ks 248 measurements in the field are commonly lower than lab Ks measurements (Nimmo et al., 2009). 249 250 This is related to a reduction in soil conductivity due to air trapping in the soil pores during the wetting process when water is applied at the land surface (Mizrahi et al., 2016; Nimmo et al., 251 252 2009).

Under the short cycles, the soil drainage process occurred mostly within the first 15 hours of the drying stage (Figure 3a). The soil drainage rate was slightly higher in summer than in winter. Under the short cycles, the model successfully followed the observed trends, where the validation period showed similar performances (Fig. S4 and Figure 3a). The model results for the short cycles confirm that the differences in drainage between summer and winter are mainly due to temperature changes that affect water viscosity (Lin et al., 2003). During the long cycle application, the drainage rates in summer showed a moderate  $\theta$  decline compared to the observed  $\theta$  under short cycles (Figure 3b). This might be due to the differences in soil physical parameters between stations 1 and 2, as highlighted by the calibrated models' parameters (Table 2). While the model under long cycles successfully followed the drainage trend during summer, the model showed poor performance during winter under long cycles. This is mainly due to the observable changes in  $\theta$  measurements, which displayed a shift towards higher values from November 2019 (Fig. S5 and Figure 2b). To explore the changes in the SAT's physical properties, an additional parameter set was calibrated against the winter data only during the long cycles (Figure 3b, green line). Both the Ks and  $\beta$  parameters attained lower values, and the  $\theta_s$ increased (Fig. S6 and Table 2). Previous studies related the accumulation of organic matter in SAT to lower rates of organic matter decomposition during winter (Nadav et al., 2012a, b; Arye et al., 2011). The authors ruled out the occurrence of soil clogging and indicated that the accumulation of organic matter at the topsoil increased the degree of soil water repellency or soil hydrophobicity. This phenomenon often develops in sandy soils (commonly used in SAT) due to the low specific surface area of sand (~0.0077 g m<sup>-2</sup>) compared to clay (~900 g m<sup>-2</sup>) (Doerr et al., 2000; Wallis and Horne, 1992). Thus, only a small amount of organic matter is required to coat the particles of the sand in order to develop soil water repellency (Wallis and Horne, 1992). Arye et al. (2011) showed that soil hydrophobicity is attributed to the reduction of liquid surface tension and increase of the contact angle. These changes in soil properties are related to the reduction of the soil permeability (Nadav et al., 2012b). It appears that long wetting and drying cycles in SAT during winter can alter the physical soil properties, which eventually affect the infiltration capabilities.

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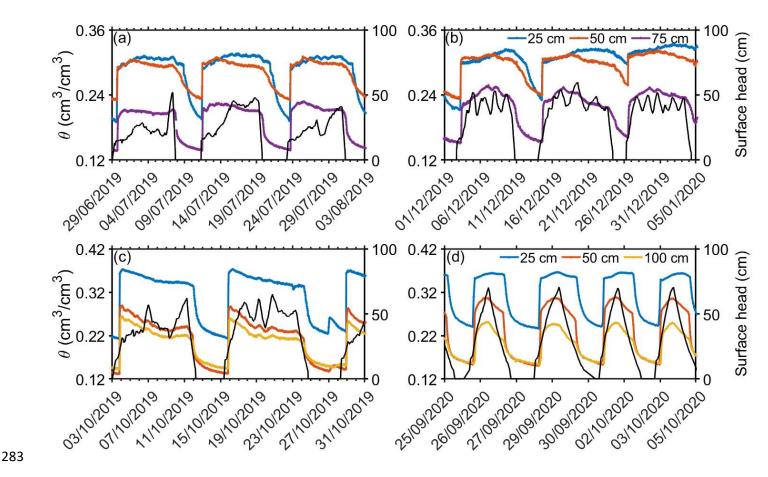


Figure 2: Representative time series of  $\theta$  measurements obtained at station 2 (Figure 1) during (a) summer and (b) winter under long wetting and drying cycles at three different depths. Black line represents the surface water hydraulic head. The bottom plots display  $\theta$  measurements obtained at station 1 under (c) long and (d) short wetting and drying cycles during the summer.

**Table 2.** Estimated parameters of the hydrological models

			Station 2
	Station 1	Station 2	(winter only)
Ks (cm h <sup>-1</sup> )	5	0.9	0.72
$ heta_s$	0.36	0.32	0.33
β	30	6.75	6.48
$ heta_{\!f\!c}$	0.19	0.19	0.19
$\psi^*$ (cm)	-15	-15	-15

 $\theta_h$  0.05 0.05

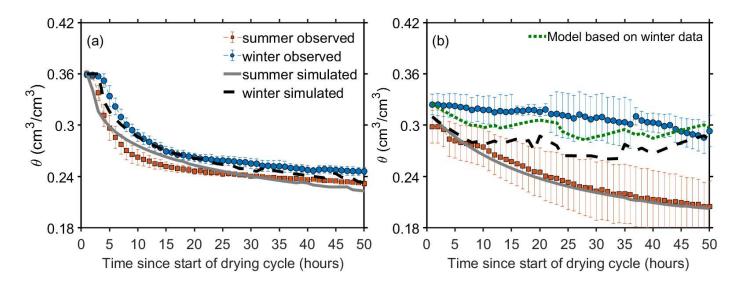


Figure 3: The average and standard deviation values of measured  $\theta$  at 25 cm depth throughout the drying stages at an hourly time scale: (a) short cycles (station 1) and (b) long cycles (station 2). The blue and red circles represent the average  $\theta$  values collected during winter (November–April) and summer (May–October), respectively. The statistics of measured  $\theta$  under long cycles are based on 19 drying stages during summer and 14 drying stages during winter. For the short cycles, the statistics are based on 12 drying stages during summer and 24 drying stages during winter. The dashed black and solid gray lines represent the average values of simulated  $\theta$  throughout the drying stage during winter (November–April) and summer (May–October), respectively. Note that for the long cycle periods, an additional model was established based only on winter data (green line).

# | Seasonal differences in SAT redox (E<sub>h</sub>) conditions under long wetting cycles

E<sub>h</sub> conditions were monitored at four depths (Figure 4a, b). The behavior of the E<sub>h</sub> dynamic following a wetting event was similar for the summer and the winter periods. At 25, 50, and 75 cm depths, a gradual decline in E<sub>h</sub> started only after a time lag from the beginning of a wetting event (Figure 4a, b). This time lag can be explained by the presence of dissolved oxygen (DO) in the percolating solution. Once DO is depleted, suboxic and anoxic conditions begin to develop

(Dutta et al., 2015; Ben Moshe et al., 2021, 2020). The E<sub>h</sub> conditions at 25 cm were the most 309 highly responsive to the wetting events, while the E<sub>h</sub> conditions were the most negative at this 310 depth (Figure 4a, b). At 100 cm depth, only minor changes were observed, and in some cases 311 (during winter), no changes were observed (Figure 4a, b). According to the gaseous O<sub>2</sub> 312 measurements (Figure 4c, d), there were partially aerated conditions (unsaturated conditions) 313 314 during some of the flooding events at 75 cm depth, while at 150 cm depth, unsaturated conditions prevailed continuously throughout the period of measurements. Therefore, the small 315 changes of E<sub>h</sub> at 100 cm depth could either have been the outcome of only minor biochemical 316 activity, or they could have been due to the sufficient oxygen supply. Gorski et al. (2019) 317 suggested that in coarse-grained soils, due to the high infiltration rates, the DO is delivered 318 rapidly by the percolating water to the base of the saturated zone. Thus, the soil microbes do not 319 320 have enough time to consume the DO. In the current study it is reflected by positive E<sub>h</sub> values at 100 cm depth (Figure 4a,b). As the wetting cycle continues, the E<sub>h</sub> conditions at 100 cm depth 321 decrease, mainly during summer (Figure 4a,b). This indicates that eventually the infiltrated water 322 that reaches 100 cm depth contains lower concentrations of DO. Yet, the E<sub>h</sub> conditions do not 323 324 show further decrease, which can be attributed to limited microbial activity. Previous studies showed that carbon availability at greater depths of the vadose zone is a dominant factor for this 325 326 limitation (Grau-martínez et al., 2018, 2017; Brettar et al., 2002). Furthermore, the monitored E<sub>h</sub> conditions illustrate that most of the activity in SAT systems occurs at the topsoil, as illustrated 327 328 in previous studies (e.g., Quanrud et al., 1996, 2003; Sopilniak et al., 2017; Grinshpan et al., 2022). Note that the E<sub>h</sub> recovery time, i.e., an increase towards positive values, was virtually 329 330 instantaneous once the drying process initiated (Figure 4a, b). The increase in E<sub>h</sub> conditions occurred concurrently with the observed rapid increase of the gaseous oxygen (O<sub>2</sub>) in the vadose 331 332 zone (Figure 4c, d). 333 A distinct difference in E<sub>h</sub> conditions between winter and summer at 25 cm depth is expressed by a more negative range of values during summer (Figure 4a, b). Since similar wastewater quality 334 and hydraulic loads were fed to the pond during summer and winter (Supporting Information), 335 the E<sub>h</sub> conditions were mainly affected by the SAT's aeration state and seasonal temperature 336 changes. In Figure 5, the monthly E<sub>h</sub> conditions at 25 cm depth are presented in the form of a 337 boxplot, together with the monthly average ambient temperature (dashed black line) and monthly 338 average global radiation (gray line). The Eh conditions showed a wider range of values with the 339

increase in temperature (Figure 5). Between November and March, the E<sub>h</sub> conditions mostly remained above zero or were slightly negative (Figure 5). Once the temperature increased above 24°C between May and October, E<sub>h</sub> conditions showed substantial fluctuation between negative and positive values (Figure 5). Note that the average monthly ambient temperatures during November and May were similar in value, but during November, the E<sub>h</sub> conditions mostly remained above zero (Figure 5). This difference is connected to the daylight duration, as indicated by the global radiation, which is substantially greater in May than in November (Figure 5, gray line). A typical characteristic of aquatic systems is the large fluctuations in dissolved oxygen (DO) concentrations due to intense photosynthesis and respiration (Stumm and Morgan, 1996). Goren et al. (2014) illustrated that following the effluent spreading in the infiltration ponds, photosynthesis enriches the water with DO. Furthermore, chemical analysis of porewater samples that were obtained at 0.5 m depth in the SAT vadose zone indicated DO's substantial influence on the biochemical conditions of the percolating water (Goren et al., 2014). Thus, the photosynthesis process enriches the effluent with DO, which encourages further microbial activity (Goren et al., 2014; Hargreaves, 2006; Rodríguez-Escales et al., 2020). However, between July and September, there was a decrease in global radiation that did not affect the E<sub>h</sub> variability (Figure 5). Thus, it appears that under long wetting stages, the seasonal temperature changes dominate the E<sub>h</sub> conditions but show some trade-off with the global radiation.

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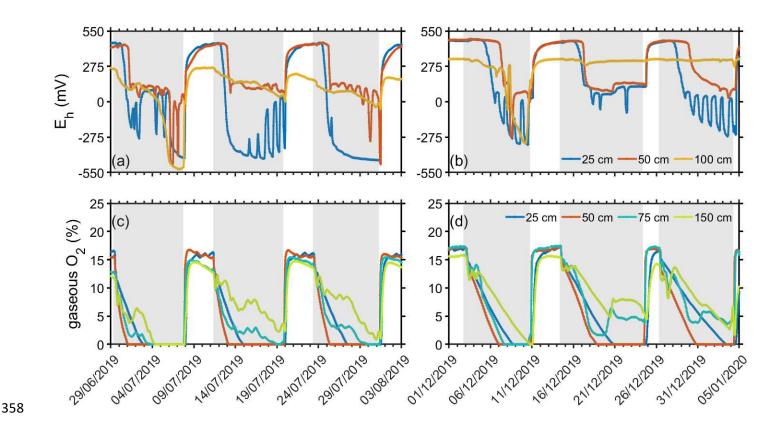


Figure 4: Representative time series of redox-potential  $(E_h)$  and gaseous  $O_2$  measurements obtained at station 2 (Figure 1) at depths of 25, 50, and 100 cm: (a, c) summer and (b, d) winter. Note that the gray and white areas indicate wetting and drying periods, respectively.

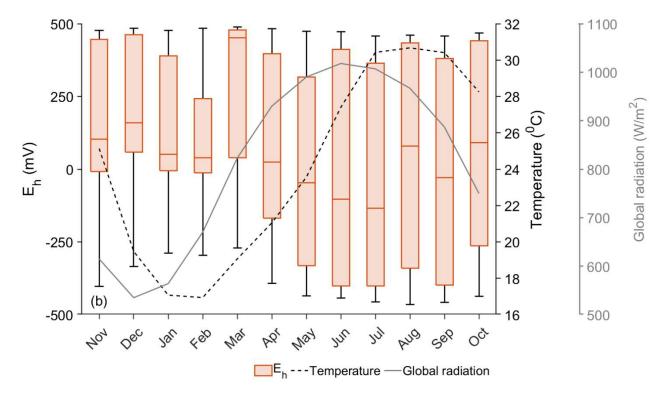


Figure 5: Seasonal changes in  $E_h$  conditions as were observed at 25 cm depth at station 2 under long (seven days) wetting periods. The dashed black line represents the monthly mean ambient temperature, and the solid gray line represents the monthly mean global radiation obtained from the Israeli Meteorological Service (IMS, 2021).

# A comparison between SAT redox (Eh) conditions under long and short cycles

It has been shown that the operational mode affects the aeration conditions of the upper vadose zone, which consequently might alter the infiltration rates and the intensity of the biogeochemical processes (Goren et al., 2014). Throughout the measurement period, the intervals of wetting and drying stages were modified (Table 1). The wetting and drying stages were substantially shortened during September 2020, which enabled examining the differences in  $E_h$  conditions under short and long cycles. The variations in  $E_h$  conditions under long cycles during October 2019, December 2019, and January 2020 are presented in Figure 6a, c. In addition, the changes in  $E_h$  conditions under short cycles during September–October 2020, December 2020, and January 2021 are presented in Figure 6b, d.

As was shown above, under long cycles, the E<sub>h</sub> conditions declined towards slightly negative values during winter and attained markedly negative values during the summer months at 25 and 50 cm depths, where only minor variations were observed at 100 cm depth (Figure 6a, c). The E<sub>h</sub> conditions, under short cycles during the summer, showed a decline towards slightly negative values for a brief time compared to long cycles (Figure 6a, b). During the winter, only minor variations in E<sub>h</sub> conditions were recorded under short cycles (Figure 6d). Note that at 100 cm depth, there was almost no change in E<sub>h</sub> conditions for either season under short cycles (Figure 6b, d). As was illustrated previously (Figure 4a, b), once the drying stage initiated, the recovery of E<sub>h</sub> towards positive values (oxic conditions) was almost instant under both short and long cycles. It appears that the re-establishment of oxic conditions occurs independently of the length of the wetting stage. The observations of E<sub>h</sub> and gaseous O<sub>2</sub> under long and short cycles indicate a weak relationship between the wetting and the drying stages. These results call into question the advantage of implementing the wet/dry ratio for optimizing SAT performance. It appears that the length of a wetting stage and a drying stage should be defined separately (Ben Moshe et al., 2020, 2021). Thus, the necessary further investigation is described below to examine the optimal lengths of the wetting and drying stages.

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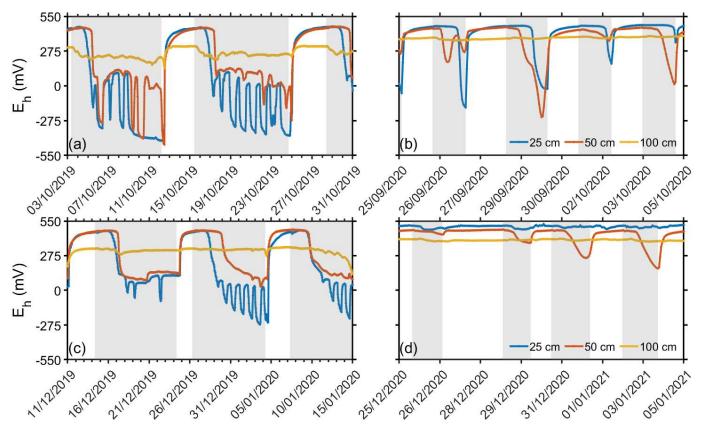


Figure 6: Representative time series of redox-potential  $(E_h)$  measurements at 25, 50 and 100 cm depths in station 1 (Figure 1): (a, c) long wetting periods and (b, d) short wetting periods. Note that the gray and white areas indicate wetting and drying periods, respectively.

# The length of the wetting stage according to E<sub>h</sub> measurements

During wetting stages, the main limiting factor of the biodegradation process is the availability of dissolved oxygen (Skopp et al., 1990; Cook and Knight, 2003). Once the soil pores are fully saturated, only the dissolved oxygen (DO) of the percolating water is available. However, studies have illustrated that the DO of the percolating water rapidly depletes (Dutta et al., 2015; Ben Moshe et al., 2021, 2020). Thus, the wetting stage should cease when the DO no longer has an effect on the degradation process, i.e., when suboxic and anoxic conditions begin to develop. Eh measurements can provide a good indication for the time when such conditions begin to be established. For this purpose, the hourly measured Eh conditions during the wetting stage of 33 recorded cycles (19 cycles during summer and 14 cycles during winter) were averaged for 25,

50, 75 and 100 cm depth (Figure 7). The data were separated between winter (November-April) 412 and summer (May-October), according to the trends presented in Figure 5. 413 During summer, the decline in E<sub>h</sub> conditions were steeper compared to the trends exhibited in 414 winter (Figure 7). Following 30 hours of wetting, the E<sub>h</sub> conditions during the summer dropped 415 416 below 400 mV at 25 cm depth, which indicates the establishment of suboxic conditions (Figure 417 7a). After 37 hours, the E<sub>h</sub> observations at 50 and 75 cm depths showed similar trends (Figure 7a). Note that after 45 hours the E<sub>h</sub> observations at 25 cm depth reach anoxic conditions. The 418 differences between E<sub>h</sub> measurements at topsoil and at deeper parts of the vadose zone were 419 previously attributed to carbon availability, which decrease with depth (Brettar et al., 2002; 420 421 Bohrerova et al., 2004). Furthermore, Brettar et al. (2002) provided a range of threshold E<sub>h</sub> values for denitrifying conditions depending on the depth of the soil horizon. Similarly, despite 422 423 the differences between the E<sub>h</sub> measurements at 25 cm depth and at deeper depths (Figure 7), all the E<sub>h</sub> measurements potentially indicate similar activity. To prevent anoxic conditions in the 424 425 SAT vadose zone during summer, the optimal length of a wetting stage, in terms of biodegradation, would be about 30 hours (1.25 days). This is in agreement with Dutta et al. 426 427 (2015), who suggested a relatively wide distribution of de-oxygenation times, between 0.36 and 428 1.5 days. In winter, the E<sub>h</sub> measurements decrease towards suboxic conditions, that occur after 51, 69 and 429 430 84 hours of wetting at 25, 50 and 75 cm depths, respectively (Figure 7b). The delay in E<sub>h</sub> drop 431 (compared to summer) is partly explained by the longer presence of gaseous O<sub>2</sub> in the SAT vadose zone (Figure 4c,d and Figure 8). It takes a longer time for the gaseous O2 to deplete 432 during winter (Figure 8), probably as a result of lower oxygen demand due to slower microbial 433 activity at low temperatures (Kirschbaum, 1995). An additional explanation for the longer 434 435 presence of gaseous O<sub>2</sub> is due to longer establishment of saturated conditions as a consequence of lower Ks values (see section Hydrological conditions). However, once the gaseous O2 is absent 436 from the SAT vadose zone during winter (Figure 8), no further decrease in E<sub>h</sub> conditions occur 437 438 (Figure 7b). Therefore, the trade-off between nutrient transport rates and reaction rates should be considered, where the supply of the percolated effluent's substrate might be faster than the 439 440 SAT's degradation capability (Greskowiak et al., 2006). Determining the length of a wetting

stage during winter, using solely the E<sub>h</sub> and gaseous O<sub>2</sub> measurements, may be challenging.

Nevertheless, the changes in soil physical parameters and the seasonal differences in E<sub>h</sub> values imply low biodegradation rates during winter. From a practical perspective, the observed E<sub>h</sub> values at 25 cm depth during winter are compared with the E<sub>h</sub> measurements during summer. Using a two-sample t-test show that the differences in the first 28 hours of a wetting cycle are insignificant. It suggests that the length of a wetting stage during winter should be no more than 30 hours, as in the summer period. Nevertheless, given the evidences that are reflected in the measurements of the current study and conclusions from previous studies, the implementation of SAT during winter is questionable. It might be necessary to store the effluents in reservoirs during winter and apply the SAT during summer. According to the E<sub>h</sub> sensor at 100 cm, suboxic conditions prevail for most times during winter and a moderate decrease (compared to the other sensors) in E<sub>h</sub> conditions is noticeable during summer (Figure 7). The gaseous O<sub>2</sub> observations at 75 cm indicate depletion of gaseous O<sub>2</sub> during summer and a very low O<sub>2</sub> concentration (2%) during winter (Figure 8c). At 150 cm, the gaseous O<sub>2</sub> observations suggest that for most of the recorded wetting cycles, unsaturated conditions prevailed (Figure 8d). It is not clear if the E<sub>h</sub> conditions at 100 cm are determined by a continuous supply of oxygen given the unsaturated conditions. This might affect the decision concerning the optimal length of a wetting cycle. However, previous studies (Miller et al., 2006; Fox et al., 2005; Lin et al., 2008; Goren et al., 2014; Sopilniak et al., 2018; Essandoh et al., 2013; Quanrud et al., 1996, 2003) and a recent study by our group (Grinshpan et al., 2022) suggest that most biodegradation activity occurs at topsoil and there is a steep reduction in removal capabilities with depth. Thus, the potential contribution of the deeper vadose zone to the SAT treatment should be further investigated.

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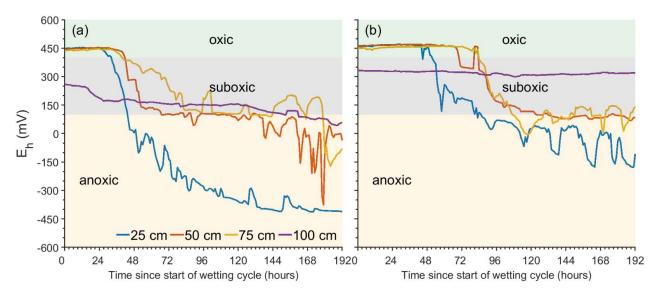
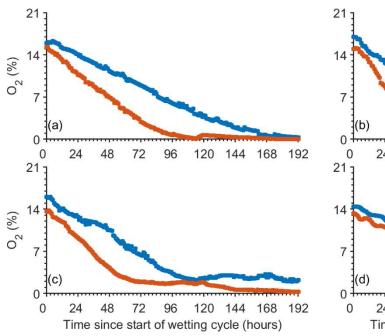


Figure 7: Plot of hourly means of  $E_h$  measurements obtained at station 2, from the beginning of the wetting stages at three depths. The statistics are based on 19 cycles during summer (May–October) and 14 cycles during winter (November–April) that were observed at (a) winter and (b) summer. Note that the data are separated between summer and winter according to the trends presented in Figure 5. The green, grey, and yellow areas represent the oxic (400mV <), suboxic  $(400mV > E_h > 100mV)$ , and anoxic  $E_h < 100mV$  conditions, respectively.



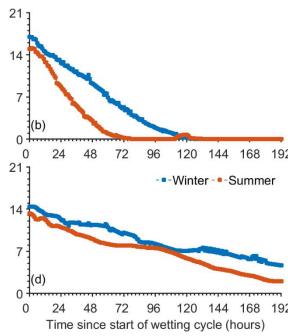


Figure 8. Plot of hourly means of gaseous  $O_2$  measurements from the beginning of the wetting stages at (a) 25, (b) 50, (c) 75 and (d) 150 cm depths. The statistics are based on 14 cycles during winter (November–April) and 19 cycles during summer (May–October) that were observed at multiple depths in station 2. Note that the data are separated between summer and winter according to the trends presented in Figure 5.

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# The length of the drying stage using $E_h$ and gaseous $O_2$ measurements

The drying stage in SAT systems is implemented to restore the infiltration, biological, and chemical capabilities of the pond, mainly by aerating the soil (Sharma and Kennedy, 2017). A drying stage is defined as the stage in which there is no water at the soil surface, and the observed volumetric water contents begin to decrease (Figure 2). To determine the optimal time for a drying stage, both the averaged values of the E<sub>h</sub> (Figure 9a,b) and gaseous O<sub>2</sub> (Figure 9c,d) observations during the drying stages are presented. Note that the E<sub>h</sub> measurements were conducted at 25, 50, 75 and 100 cm depths and the O<sub>2</sub> observations were obtained at 25, 50, 75 and 150 cm depths (Figure 9). The E<sub>h</sub> and O<sub>2</sub> data during the drying stages were separated between winter (November-April) and summer (May-October), according to the trends presented in Figure 5. Slower aeration rates and larger variability were observed during winter compared to summer (Figure 9a,b). Furthermore, the E<sub>h</sub> recovery is clearly dominated by the rates of the gaseous O<sub>2</sub> intrusion to the soil (Figure 9c,d). During winter, the aeration rates are different at each depth, but in general the gaseous O<sub>2</sub> concentrations increase moderately with time (Figure 9a). The recovery of the E<sub>h</sub> conditions display comparable trends (Figure 9c). Although the O<sub>2</sub> sensor at 50 cm depth suggest a faster O<sub>2</sub> intrusion rate compared to the other sensors (Figure 9a), it is not expressed in the E<sub>h</sub> recovery time (Figure 9c). During summer all the observed O<sub>2</sub> curves show steep recovery that ceases after about 20 hours (Figure 9b). According to the O<sub>2</sub> observations, the gaseous O<sub>2</sub> intrusion process at 50 and 75 cm depths starts at the very beginning of the drying stage, while at 25 and 100 cm depths there is a two-hour delay (Figure 9b). Nevertheless, the E<sub>h</sub> conditions recovery is comparable at all depths (Figure 9d).

The E<sub>h</sub> observations illustrate that the re-establishment of oxic conditions in the SAT vadose zone is similar during winter and summer (Figure 9c,d). Once the drying stage started, it required about 36 hours for the E<sub>h</sub> conditions to reach values in the range of oxic conditions, regardless of the initial value of the E<sub>h</sub> conditions (Figure 9c,d). However, a recent study has indicated that using solely the gaseous O<sub>2</sub> concentration to quantify soil aeration status might be inaccurate for some conditions (Ben-Noah et al., 2021). Instead, the authors suggested using the Damkholer number, which is the ratio between characteristic diffusion (i.e., oxygen supply) and soil respiration times. A small Damkholer number would indicate that the O<sub>2</sub> consumption rate is lower than the O<sub>2</sub> supply. Thus, although during summer the oxygen supply rate is relatively high compared to winter (Error! Reference source not found.a,b), the oxygen consumption rate for soil respiration is expected to be substantially higher during summer than during winter (Kirschbaum, 1995), as it is reflected by the E<sub>h</sub> observations (Figure 5 and Figure 7). Therefore, in parallel to proper treatment of aeration time, optimal wetting time should be further treated.

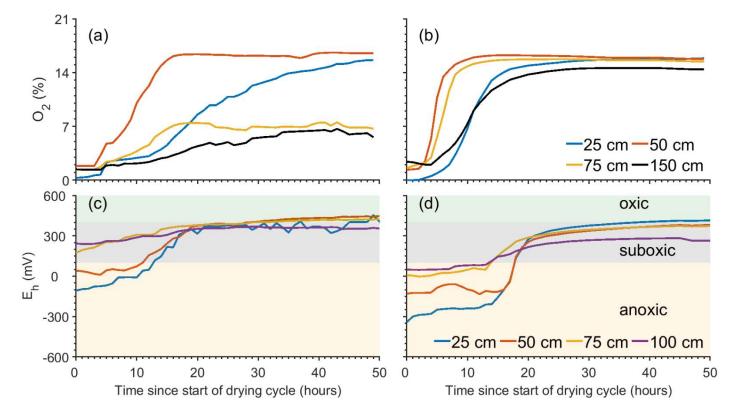


Figure 9: Plot of hourly means of  $E_h$  and gaseous  $O_2$  measurements from the beginning of the drying stages. The statistics are based on (a,c) 14 cycles during winter (November–April) and (b,d) 19 cycles during summer (May–October). Note that the gaseous  $O_2$  (a,b) measurements were obtained at 25, 50, 75 and 150 cm depths and the  $E_h$  conditions (c,d) were observed at 25, 50, 75 and 100 cm depths. All observations were conducted in station 2. The data are separated between summer and winter according to the trends presented in Figure 5. The green, grey, and yellow areas represent the oxic  $(400 \text{mV} < E_h > 100 \text{mV})$ , and anoxic  $E_h < 100 \text{mV}$  conditions, respectively.

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## **4 Summary and Conclusions**

Continuous monitoring of  $E_h$ ,  $\theta$ , T, and gaseous  $O_2$  in the vadose zones of SAT infiltration ponds was carried out for about 600 days. SAT operation was subjected to long and short wetting and drying cycles and seasonal changes. The datasets enabled the examination of factors that control the hydrological and geochemical conditions in SAT. Calibrated and validated hydrological models were applied to investigate the water flow dynamics in the SAT vadose zone. E<sub>h</sub> and gaseous O<sub>2</sub> observations were averaged to determine seasonal changes and deduce the optimal length of wetting and drying stages. The examination of the measured E<sub>h</sub> conditions illustrated a noticeable decline to markedly negative values (-450mV >) during summer and to values between 0 and -50 mV during winter. These E<sub>h</sub> conditions were established following 30 hours of wetting, and no considerable changes in E<sub>h</sub> conditions were noticeable until the wetting stages ceased. A monthly statistic of the E<sub>h</sub> measurements illustrated the relationship between the size of the E<sub>h</sub> amplitude and the seasonal T changes. Furthermore, it is speculated that the limited decrease in E<sub>h</sub> conditions during winter were due to lower microbial activity. An additional support for this claim is the reduction in infiltration capabilities following long wetting and drying cycles during winter, as was indicated by the hydrological models. To define the optimal length of a wetting stage, the E<sub>h</sub> data were averaged and separated between the winter and summer periods. During the summer period, the optimal time length of a wetting stage, according to E<sub>h</sub> observations, is about 30 hours. Determining the optimal length of a wetting stage during winter is challenging. Practically, there are no significant differences between E<sub>h</sub> conditions during winter and summer

for the first 28 hours of wetting. Thus, the length of a wetting stage during winter should be 30 547 hours, as in the summer period. 548 The length of a drying stage, following the E<sub>h</sub> observations, should be about 36 hours, regardless 549 of the initial values of the E<sub>h</sub> conditions and the season. Note that during summer, a longer drying 550 551 time is required for the E<sub>h</sub> conditions to attain suboxic conditions. Nevertheless, the gaseous O<sub>2</sub> observations indicated faster aeration rates during summer, which compensate for the very 552 negative E<sub>h</sub> conditions and allow fast recovery. Following our analysis, we suggest that under the 553 tested conditions, the length of a drying stage should be 36 hours for winter and summer. Shorter 554 drying stages would affect SAT efficiency, while applying longer drying stages would reduce the 555 556 total hydraulic loads that can be fed to the infiltration basin. 557 Implementing in situ  $E_h$ ,  $\theta$ , T and gaseous  $O_2$  sensors provided continuous high-resolution observations. These datasets revealed the hydrological and biochemical dynamics in SAT as 558 imposed by seasonal and operational changes. Analysis of the E<sub>h</sub> and O<sub>2</sub> measurements enabled 559 560 the identification of the optimal time lengths of wetting and drying stages. The results indicated that there is no direct relationship between the length of a wetting stage and a drying stage. Thus, 561 the operational use of a wetting/drying ratio in SAT management should be reconsidered. 562 563 Acknowledgments 564 This work was financed within the framework of the German–Israeli Water Technology 565 Cooperation Program under Project No. WT1601/2689, by the German Federal Ministry of 566 567 Education and Research (BMBF), and by the Israel Ministry of Science, Technology and Space

(MOST).

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