1	Continuous Monitoring of a Soil Aquifer Treatment System's Physico-Chemical			
2	Conditions to Optimize Operational Performance			
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13	Highlights			
14	• Long wetting stages reduce soil percolation capabilities during winter.			
15	• Redox and gaseous O_2 display intensive dynamics in the top 25 cm of the soil aquifer			
16	treatment vadose zone.			
17	• Optimal wetting and drying stages are defined according to E_h and gaseous O_2			
18	observations.			
19	• The length of wetting and drying stages should be defined separately rather than by			
20	adhering to their ratio.			
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22				

24 Abstract

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_h) was continuously monitored, together with other variables such as volumetric water content (θ), soil temperature, and gaseous oxygen 33 (O₂), 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_h observations, under long wetting stages, demonstrated 38 39 larger variability and very negative values as ambient temperature increased. Assembling the daily E_h 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_h and O₂ 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

47 **1 Introduction**

Worldwide water scarcity has motivated the development of alternative water resources such as 48 the reuse of treated wastewater. Soil aquifer 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

59 Kennedy, 2017).

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 O₂ (Ben Moshe et al., 2020; Massmann et al., 2006). 70

A consequence of the perturbation in the O₂ supply to SAT is expressed in changes in the redox conditions (Mächler et al., 2013; Rezanezhad et al., 2014). Redox potential or oxidationreduction potential is a quantitative measure of electron availability, i.e., the tendency of the system to receive or donate electrons (Hinchey and Schaffner, 2005). Substantial changes in SAT systems' redox conditions might lead to the release of undesirable metals, such Fe²⁺ and

 Mn^{2+} (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 O₂/H₂O, 81 NO_3/N_2 , MnO_2/Mn^{2+} , Fe^{3+}/Fe^{2+} , and SO_4/H_2S , 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_2) is depleted, the next strongest 85 oxidizing species is used (NO₃) 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 O₂ 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 (E_h) , together with chemical and physical parameters such as 106 water content, soil temperature, O₂ concentration, etc., can be continuously monitored by 107 108 installing the relevant sensors. Previous studies have implemented the E_h sensor and successfully described, with high temporal resolution, the subsurface chemical conditions in various 109 environments, such as wetlands, the groundwater (or the capillary fringe), aquifers, etc. (Wallace 110 et al., 2019; McMahon and Chapelle, 2008; Shenker et al., 2005; Silver et al., 2018; Rezanezhad 111 et al., 2014). To improve SAT system performances, the link between the wetting and drying 112 stages and the subsequent redox conditions developed in the subsurface should be established. 113 Thus, in situ monitoring can improve SAT management performance and reduce the subjectivity 114 of the operator. The objective of this study was to examine the temporal variability in redox 115 potential and the way it is affected by changes in volumetric water content, gaseous O_2 , and 116 climate imposed by different operational modes of wetting and drying stages. Furthermore, 117 calibrated and validated hydrological models were used to explore the behavior of water fluxes 118 119 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. 120

121

122 **2** Methods

123 Study sites

The Dan region reclamation project (Shafdan) reclaims about 125 million m³ of effluent 124 annually, from the Tel Aviv metropolitan area in Israel. The treatment of effluents occurs in two 125 stages. The first stage involves mechanical-biological treatment, which is based on activated 126 sludge, while in the second stage, the treated water (a secondary effluent) is delivered to 127 128 infiltration basins, as part of the SAT system, to further improve water quality. Six infiltration basin sites, covering a total area of 1.053 km^2 , are located in central Israel, overlying the coastal 129 aquifer (Figure 1). Each basin is divided into several spreading ponds, about 1500 m² each, 130 which are alternately flooded. The vadose zone that underlies the basins is mostly composed of 131 sand, sandy loam soil, and calcareous sandstone layers. Typically, the ponds are flooded for one 132 to two days (max hydraulic head of about 50 cm), followed by two to six days of drainage and 133 soil surface drying. The wetting and drying stages are controlled by the ponds' flooding order, 134

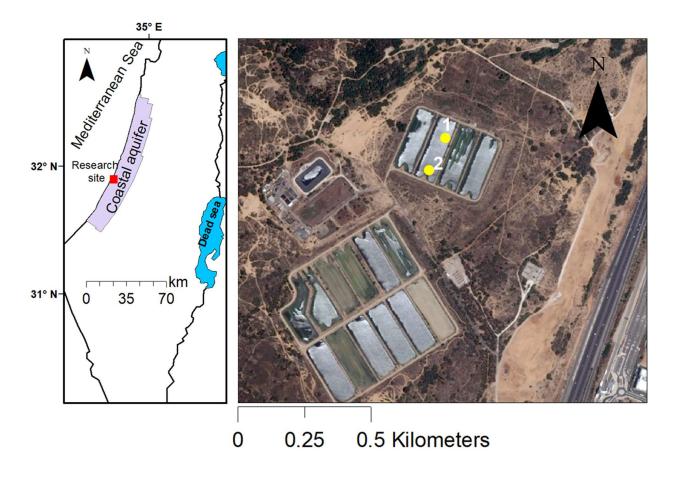
- the availability of effluent, and the drying period, which is suggested to be at least 24 h (Icekson-
- 136 Tal et al., 2003). The basin surface is plowed on a regular basis to break up the developed
- 137 biocrust and to prevent clogging (see Negev et al. (2020) for details).

138 Study operation

- 139 In this research, two in situ measurement stations were installed in an infiltration pond during
- 140 2018 (pond 4103 in the Yavne 1 cluster, Figure 1). Each station was equipped with several sets
- of sensors at 25, 50, 75, 100 and 150 cm depth, including time domain transmittance (TDT)
- 142 probes (Acclima Inc., Idaho, USA), copper-constantan thermocouples (OMEGA Engineering,
- 143 Inc., Connecticut, USA), oxidation-reduction potential (ORP) electrodes (ELH016 van London
- 144 Co, Houston, TX, USA), and O₂ percentage probes (ICT02 sensor, ICT Int., Australia). Data
- 145 were collected at prescribed intervals and logged on a CR1000 data logger (Campbell Scientific,
- 146 Inc., Logan, UT, USA). In addition, suction cups were installed at similar depths. In station 1
- 147 (Figure 1), the data consisted of volumetric water content (θ), soil temperature (T), and ORP (E_h)
- time series, which were continuously measured every 20 minutes between 28/07/2020 and
- 149 10/02/2021 (total of 14,185 values, 197 days, for each variable). The data were obtained at 25,
- 150 50, and 100 cm depths. In station 2 (Figure 1), θ , T, gaseous oxygen (O₂), and E_h were measured
- every 20 minutes between 08/05/2019 and 20/07/2020. There were about 60 days in which data
- were not collected in station 2 due to technical issues. The data from station 2 contained 27,222
- points of θ , 29,394 points of O₂, 30,414 points of E_h, and 26,730 points of T measurements. In
- station 2, the data were collected at 25, 50, 75 and 100 cm depths.

155 The water quality characteristics of the secondary effluent that flooded the Yavne 1 basin are

- 156 presented in the *Supporting Information* (Fig. S1). Note that the quality parameter concentrations
- 157 conform to the updated "Inbar" regulations (Inbar, 2007) and the findings of a previous study
- 158 that surveyed numerous wastewater storage and treatment reservoirs across the country (Kfir et
- al., 2012). To determine the soil physical properties, undisturbed soil cores were sampled at
- 160 different depths. Subsequently, flow experiments were carried out to calculate the saturated
- 161 hydraulic conductivity (*Ks*) based on Darcy's Law (*Supporting Information*, Fig. S2).
- 162 Additionally, particle size distribution (PSD) analyses are presented in Fig. S3 (*Supporting*
- 163 *Information*). The PSD results indicate that the SAT vadose zone is homogeneous.



164

165 Figure 1: *The location of the investigated site, the Yavne 1 infiltration basin of the Shafdan.*

166 In the close up of the investigated pond, the yellow circles represent the locations of the

167 *measurement stations (*©*Google Earth)*.

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169 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):

176
$$q = Ks \times \frac{(L+d-\psi^*)}{L} \quad (1)$$

177 where
$$\frac{q}{L}$$
 (L T⁻¹) is the water flux, $\frac{d}{L}$ (L) is the ponding depth, $\frac{L}{L}$ (L) is the thickness of the

saturated vadose zone, and ψ^* (L) is the negative pressure head at the wetting front. Note that L

179 is assumed here to be constant, the subsurface is assumed to be homogeneous, and θ is assumed

to vary with time only. As the wetting front progresses, the gradient approaches a value of unity,

181 and the infiltration rate becomes equal to the hydraulic conductivity of the wetted zone. Once the

182 water ponding ceases, the water drainage is set equal to the unsaturated hydraulic conductivity,

183 described with an exponential form (Guswa et al., 2002):

184
$$D(\theta) = Ks \frac{e^{\beta(\theta - \theta_{fc})} - 1}{e^{\beta(\theta_s - \theta_{fc})} - 1}$$
(2)

185 where K_s (L T⁻¹) is the saturated hydraulic conductivity, β is a parameter of the soil, θ (L³ L⁻³) is 186 the volumetric water content, θ_{fc} (L³ L⁻³) is the water content at field capacity, and θ_s (L³ L⁻³) is 187 the saturated water content. Furthermore, the effect of temperature changes on the soil hydraulic 188 conductivity is implemented through the change in viscosity (Lin et al., 2003):

189
$$Ks_T = Ks_{25} \frac{\mu_{25}}{\mu_T}$$
(3)

where Ks_T and Ks_{25} are soil hydraulic conductivity values at temperature T°C and 25°C, respectively, and μ_T and μ_{25} are the dynamic viscosity of water (M L⁻¹ T⁻¹) values at T°C and 25° C, respectively.

193 An inverse problem was set to find an optimal combination of $\frac{Ks}{\delta}$ and $\frac{\beta}{\delta}$ parameters that

194 minimizes the following objective function:

195
$$\Phi(b) = \sum_{i=1}^{N} [\theta(t_i) - \theta(t_i, b)]^2 \quad (4)$$

- 196 where N is the number of the θ observations, $\theta(t_i)$ are the observations at a specific time, and 197 $\theta(t_i,b)$ are the corresponding models' (Eq. 1 to 3) predictions for the vector of optimized 198 parameters, b (*Ks* and β). The inverse problem was solved using the *fminsearch* function in
- 199 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).

202

203 **3 Results and Discussion**

- 204 The water level measurements at the soil surface were collected by the Shafdan operators as part
- 205 of the operational routine. Analysis of the water level data (ponding depth) indicates two
- 206 operational modes of long and short cycles that were implemented at the investigated pond. The
- 207 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
- 209 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
- 211 exist due to microtopography and the distance between the inlet and the far parts of the pond.
- 212 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
- 215 Mediterranean climate. Our monitoring systems are operated independently from the Shafdan
- 216 facilities.

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

Long cycles Short cycles Wetting stage (days) 9 ± 2.4 1.5 ± 0.4 Drying stage (days) 3.3 ± 2.4 1.8 ± 1 Number of recorded cycles 37 33 Length of cycle (days) 12.7 ± 3 3.2 ± 1.3 Wet/dry ratio 3 ± 1.8 0.9 ± 0.3

220

221 Hydrological conditions

A representative set of θ 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 θ 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 (Figure 2b). The water content variations under short cycles were measured at station 1 (Figure 227 $\frac{2}{2}$, 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, θ measurements were obtained throughout 19 cycles during summer 230 (May-October) and 14 cycles during winter (November-April). Under short cycles, there were 231

232 12 cycles during summer and 25 during winter.

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 β parameters (Fig. S4 and Fig. S5, Supporting Information, Table 2). Throughout the calibration, the Ks and β 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).

- 253 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.
- 255 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 θ decline compared to the observed
- 260 θ 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).
- 262 While the model under long cycles successfully followed the drainage trend during summer, the
- 263 model showed poor performance during winter under long cycles. This is mainly due to the
- 264 observable changes in θ measurements, which displayed a shift towards higher values from
- 265 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
- 267 cycles (Figure 3b, green line). Both the Ks and β parameters attained lower values, and the θ_s
- increased (Fig. S6 and Table 2). Previous studies related the accumulation of organic matter in
- 269 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
- 272 hydrophobicity. This phenomenon often develops in sandy soils (commonly used in SAT) due to
- the low specific surface area of sand ($\sim 0.0077 \text{ g m}^{-2}$) compared to clay ($\sim 900 \text{ g m}^{-2}$) (Doerr et al.,
- 274 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
- 278 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
- 280 infiltration capabilities.
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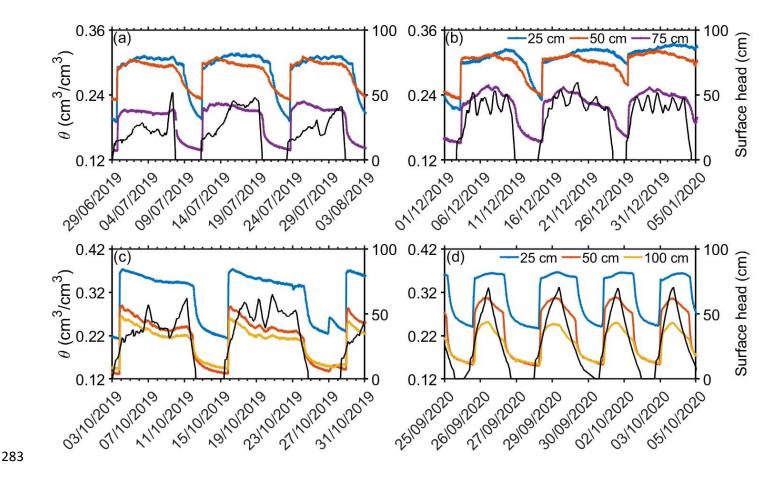


Figure 2: Representative time series of θ measurements obtained at station 2 (Figure 1) during (a) summer and (b) winter under long wetting and drying cycles at three different depths. Black line represents the surface water hydraulic head. The bottom plots display θ measurements obtained at station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

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Table 2. Estimated parameters of the hydrological models

			Station 2
	Station 1	Station 2	(winter only)
Ks (cm h ⁻¹)	5	0.9	0.72
$ heta_s$	0.36	0.32	0.33
β	30	6.75	6.48
$ heta_{fc}$	0.19	0.19	0.19
ψ^* (cm)	-15	-15	-15



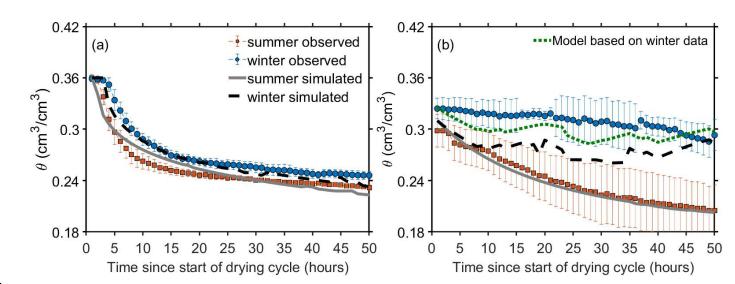




Figure 3: The average and standard deviation values of measured $\frac{\theta}{\theta}$ at 25 cm depth throughout 292 the drying stages at an hourly time scale: (a) short cycles (station 1) and (b) long cycles (station 293 2). The blue and red circles represent the average $\frac{\theta}{\theta}$ values collected during winter (November– 294 April) and summer (May–October), respectively. The statistics of measured $\frac{\theta}{\theta}$ under long cycles 295 are based on 19 drying stages during summer and 14 drying stages during winter. For the short 296 cycles, the statistics are based on 12 drying stages during summer and 24 drying stages during 297 298 winter. The dashed black and solid gray lines represent the average values of simulated $\frac{\theta}{\theta}$ 299 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 300

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only on winter data (green line).

303 Seasonal differences in SAT redox (E_h) conditions under long wetting cycles

E_h conditions were monitored at four depths (Figure 4a, b). The behavior of the E_h 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_h 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_h conditions at 25 cm were the most 309 highly responsive to the wetting events, while the $E_{\rm h}$ 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_2 312 measurements (Figure 4c, d), there were partially aerated conditions (unsaturated conditions) 313 during some of the flooding events at 75 cm depth, while at 150 cm depth, unsaturated 314 conditions prevailed continuously throughout the period of measurements. Therefore, the small 315 changes of E_h 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 have enough time to consume the DO. In the current study it is reflected by positive E_h values at 320 100 cm depth (Figure 4a,b). As the wetting cycle continues, the $E_{\rm h}$ 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_h conditions do not 323 show further decrease, which can be attributed to limited microbial activity. Previous studies 324 showed that carbon availability at greater depths of the vadose zone is a dominant factor for this 325 limitation (Grau-martínez et al., 2018, 2017; Brettar et al., 2002). Furthermore, the monitored E_{h} 326 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_h 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_h conditions 331 occurred concurrently with the observed rapid increase of the gaseous oxygen (O_2) in the vadose 332 zone (Figure 4c, d).

A distinct difference in E_h 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 and hydraulic loads were fed to the pond during summer and winter (*Supporting Information*), the E_h conditions were mainly affected by the SAT's aeration state and seasonal temperature changes. In Figure 5, the monthly E_h conditions at 25 cm depth are presented in the form of a boxplot, together with the monthly average ambient temperature (dashed black line) and monthly average global radiation (gray line). The E_h conditions showed a wider range of values with the

increase in temperature (Figure 5). Between November and March, the E_h conditions mostly 340 remained above zero or were slightly negative (Figure 5). Once the temperature increased above 341 342 24°C between May and October, E_h conditions showed substantial fluctuation between negative and positive values (Figure 5). Note that the average monthly ambient temperatures during 343 November and May were similar in value, but during November, the E_h conditions mostly 344 remained above zero (Figure 5). This difference is connected to the daylight duration, as 345 indicated by the global radiation, which is substantially greater in May than in November (Figure 346 5, gray line). A typical characteristic of aquatic systems is the large fluctuations in dissolved 347 oxygen (DO) concentrations due to intense photosynthesis and respiration (Stumm and Morgan, 348 **1996**). Goren et al. (2014) illustrated that following the effluent spreading in the infiltration 349 ponds, photosynthesis enriches the water with DO. Furthermore, chemical analysis of porewater 350 samples that were obtained at 0.5 m depth in the SAT vadose zone indicated DO's substantial 351 influence on the biochemical conditions of the percolating water (Goren et al., 2014). Thus, the 352 photosynthesis process enriches the effluent with DO, which encourages further microbial 353 activity (Goren et al., 2014; Hargreaves, 2006; Rodríguez-Escales et al., 2020). However, 354 355 between July and September, there was a decrease in global radiation that did not affect the $E_{\rm h}$ variability (Figure 5). Thus, it appears that under long wetting stages, the seasonal temperature 356 357 changes dominate the E_h conditions but show some trade-off with the global radiation.

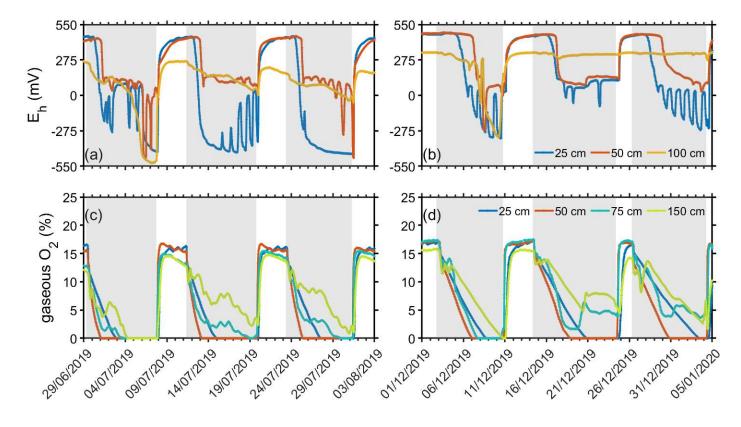


Figure 4: Representative time series of redox-potential (E_h) and gaseous O_2 measurements

- 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.*

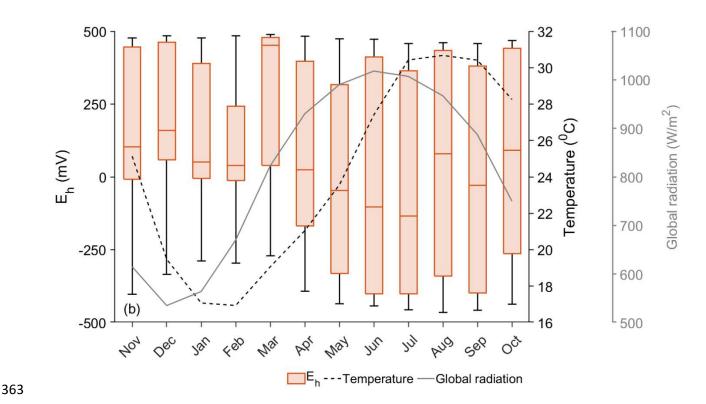


Figure 5: Seasonal changes in *E_h* conditions as were observed at 25 cm depth at station 2 under long (seven days) wetting periods. The dashed black line represents the monthly mean ambient temperature, and the solid gray line represents the monthly mean global radiation obtained from the Israeli Meteorological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

369 A comparison between SAT redox (E_h) conditions under long and short cycles

370 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

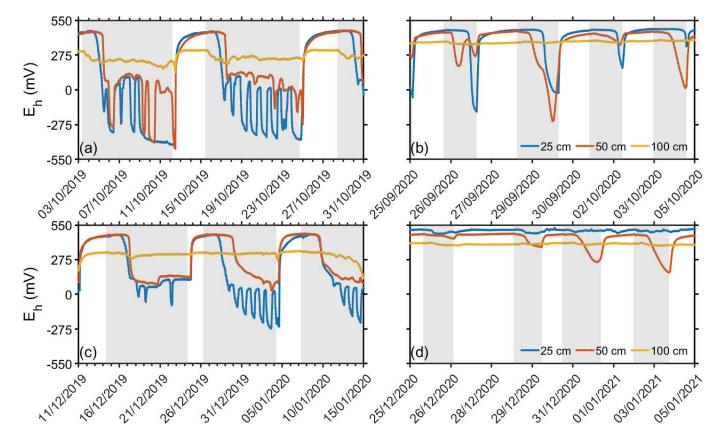
biogeochemical processes (Goren et al., 2014). Throughout the measurement period, the

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,
- 378 December 2020, and January 2021 are presented in Figure 6b, d.

As was shown above, under long cycles, the E_h conditions declined towards slightly negative 379 values during winter and attained markedly negative values during the summer months at 25 and 380 381 50 cm depths, where only minor variations were observed at 100 cm depth (Figure 6a, c). The E_h conditions, under short cycles during the summer, showed a decline towards slightly negative 382 values for a brief time compared to long cycles (Figure 6a, b). During the winter, only minor 383 variations in E_h conditions were recorded under short cycles (Figure 6d). Note that at 100 cm 384 depth, there was almost no change in E_h conditions for either season under short cycles (Figure 385 6b, d). As was illustrated previously (Figure 4a, b), once the drying stage initiated, the recovery 386 of E_h towards positive values (oxic conditions) was almost instant under both short and long 387 cycles. It appears that the re-establishment of oxic conditions occurs independently of the length 388 of the wetting stage. 389

The observations of E_h and gaseous O_2 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.



397

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.

402 The length of the wetting stage according to E_h measurements

403 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 404 405 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 406 407 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. $E_{\rm h}$ 408 409 measurements can provide a good indication for the time when such conditions begin to be established. For this purpose, the hourly measured E_h conditions during the wetting stage of 33 410 411 recorded cycles (19 cycles during summer and 14 cycles during winter) were averaged for 25,

412 50, 75 and 100 cm depth (Figure 7). The data were separated between winter (November–April)
413 and summer (May–October), according to the trends presented in Figure 5.

414 During summer, the decline in E_h conditions were steeper compared to the trends exhibited in

415 winter (Figure 7). Following 30 hours of wetting, the $E_{\rm h}$ conditions during the summer dropped

416 below 400 mV at 25 cm depth, which indicates the establishment of suboxic conditions (Figure

- 417 7a). After 37 hours, the E_h observations at 50 and 75 cm depths showed similar trends (Figure
- 418 7a). Note that after 45 hours the E_h observations at 25 cm depth reach anoxic conditions. The
- 419 differences between E_h measurements at topsoil and at deeper parts of the vadose zone were
- 420 previously attributed to carbon availability, which decrease with depth (Brettar et al., 2002;
- 421 Bohrerova et al., 2004). Furthermore, Brettar et al. (2002) provided a range of threshold E_h
- 422 values for denitrifying conditions depending on the depth of the soil horizon. Similarly, despite
- 423 the differences between the E_h measurements at 25 cm depth and at deeper depths (Figure 7), all

424 the E_h measurements potentially indicate similar activity. To prevent anoxic conditions in the

425 SAT vadose zone during summer, the optimal length of a wetting stage, in terms of

- 426 biodegradation, would be about 30 hours (1.25 days). This is in agreement with Dutta et al.
- 427 (2015), who suggested a relatively wide distribution of de-oxygenation times, between 0.36 and
- 428 **1.5 day**s.
- 429 In winter, the E_h measurements decrease towards suboxic conditions, that occur after 51, 69 and
- 430 84 hours of wetting at 25, 50 and 75 cm depths, respectively (Figure 7b). The delay in E_h drop

431 (compared to summer) is partly explained by the longer presence of gaseous O_2 in the SAT

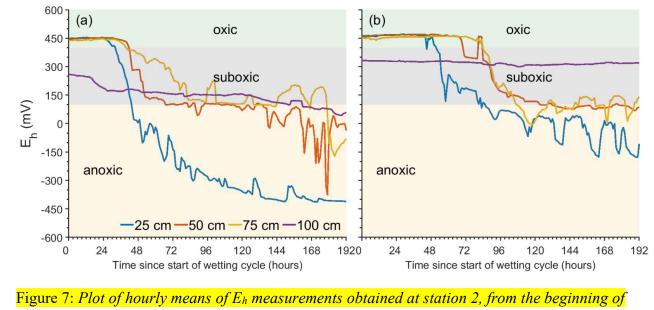
- 432 vadose zone (Figure 4c,d and Figure 8). It takes a longer time for the gaseous O₂ to deplete
- 433 during winter (Figure 8), probably as a result of lower oxygen demand due to slower microbial
- 434 activity at low temperatures (Kirschbaum, 1995). An additional explanation for the longer

435 presence of gaseous O₂ is due to longer establishment of saturated conditions as a consequence of

436 lower Ks values (see section *Hydrological conditions*). However, once the gaseous O₂ is absent

- 437 from the SAT vadose zone during winter (Figure 8), no further decrease in E_h conditions occur
- 438 (Figure 7b). Therefore, the trade-off between nutrient transport rates and reaction rates should be
- 439 considered, where the supply of the percolated effluent's substrate might be faster than the
- 440 SAT's degradation capability (Greskowiak et al., 2006). Determining the length of a wetting
- 441 stage during winter, using solely the E_h and gaseous O_2 measurements, may be challenging.

- 442 Nevertheless, the changes in soil physical parameters and the seasonal differences in E_h values
- 443 imply low biodegradation rates during winter. From a practical perspective, the observed E_h
- 444 values at 25 cm depth during winter are compared with the E_h measurements during summer.
- 445 Using a two-sample t-test show that the differences in the first 28 hours of a wetting cycle are
- 446 insignificant. It suggests that the length of a wetting stage during winter should be no more than
- 447 30 hours, as in the summer period. Nevertheless, given the evidences that are reflected in the
- 448 measurements of the current study and conclusions from previous studies, the implementation of
- 449 SAT during winter is questionable. It might be necessary to store the effluents in reservoirs
- 450 during winter and apply the SAT during summer.
- 451 According to the E_h sensor at 100 cm, suboxic conditions prevail for most times during winter
- 452 and a moderate decrease (compared to the other sensors) in E_h conditions is noticeable during
- 453 summer (Figure 7). The gaseous O_2 observations at 75 cm indicate depletion of gaseous O_2
- 454 during summer and a very low O_2 concentration (2%) during winter (Figure 8c). At 150 cm, the
- 455 gaseous O₂ observations suggest that for most of the recorded wetting cycles, unsaturated
- 456 conditions prevailed (Figure 8d). It is not clear if the E_h conditions at 100 cm are determined by a
- 457 continuous supply of oxygen given the unsaturated conditions. This might affect the decision
- 458 concerning the optimal length of a wetting cycle. However, previous studies (Miller et al., 2006;
- 459 Fox et al., 2005; Lin et al., 2008; Goren et al., 2014; Sopilniak et al., 2018; Essandoh et al., 2013;
- 460 Quanrud et al., 1996, 2003) and a recent study by our group (Grinshpan et al., 2022) suggest that
- 461 most biodegradation activity occurs at topsoil and there is a steep reduction in removal
- 462 capabilities with depth. Thus, the potential contribution of the deeper vadose zone to the SAT
- 463 treatment should be further investigated.



467 the wetting stages at three depths. The statistics are based on 19 cycles during summer (May-

468 October) and 14 cycles during winter (November–April) that were observed at (a) winter and (b)
469 summer. Note that the data are separated between summer and winter according to the trends

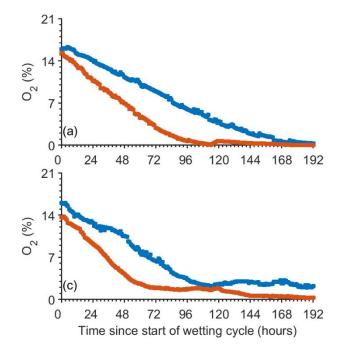
470 presented in Figure 5. The green, grey, and yellow areas represent the oxic (400mV <), suboxic

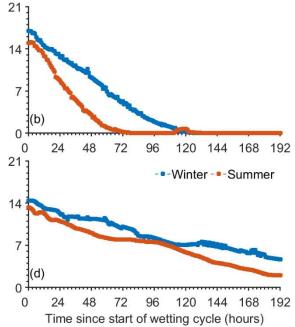
471 (400mV > E_h > 100mV), and anoxic E_h < 100mV) conditions, respectively.

472

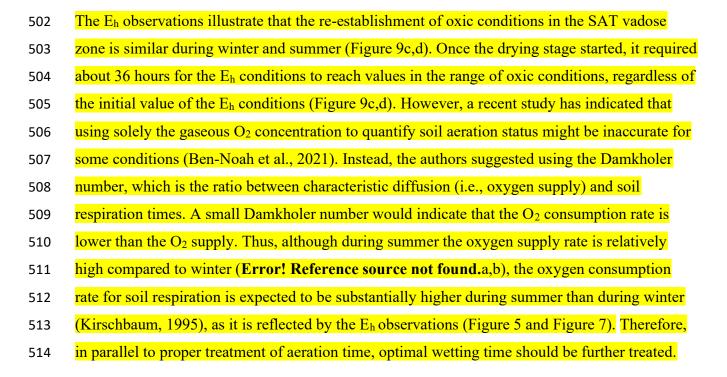
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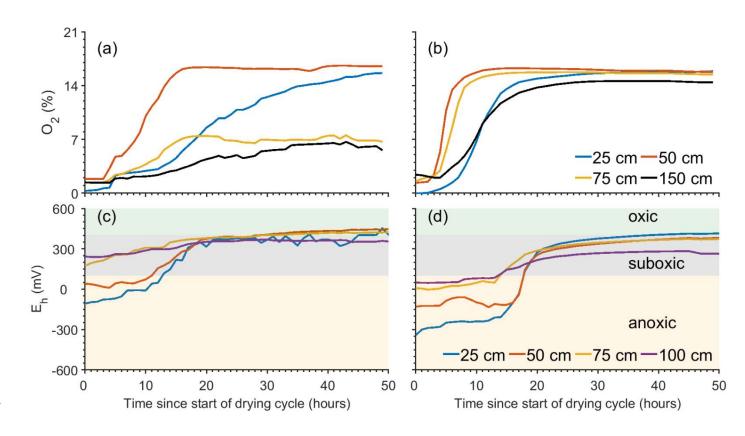
466





474	Figure 8. Plot of hourly means of gaseous O_2 measurements from the beginning of the wetting
475	stages at (a) 25, (b) 50, (c) 75 and (d) 150 cm depths. The statistics are based on 14 cycles
476	during winter (November–April) and 19 cycles during summer (May–October) that were
477	observed at multiple depths in station 2. Note that the data are separated between summer and
478	winter according to the trends presented in Figure 5.
479	
480	The length of the drying stage using E_h and gaseous O_2 measurements
481	The drying stage in SAT systems is implemented to restore the infiltration, biological, and
482	chemical capabilities of the pond, mainly by aerating the soil (Sharma and Kennedy, 2017). A
483	drying stage is defined as the stage in which there is no water at the soil surface, and the
484	observed volumetric water contents begin to decrease (Figure 2). To determine the optimal time
485	for a drying stage, both the averaged values of the E_h (Figure 9a,b) and gaseous O ₂ (Figure 9c,d)
486	observations during the drying stages are presented. Note that the E_h measurements were
487	conducted at 25, 50, 75 and 100 cm depths and the O_2 observations were obtained at 25, 50, 75
488	and 150 cm depths (Figure 9). The E_h and O_2 data during the drying stages were separated
489	between winter (November–April) and summer (May–October), according to the trends
490	presented in Figure 5.
491	Slower aeration rates and larger variability were observed during winter compared to summer
492	(Figure 9a,b). Furthermore, the E_h recovery is clearly dominated by the rates of the gaseous O_2
493	intrusion to the soil (Figure 9c,d). During winter, the aeration rates are different at each depth,
494	but in general the gaseous O_2 concentrations increase moderately with time (Figure 9a). The
495	recovery of the E_h conditions display comparable trends (Figure 9c). Although the O ₂ sensor at
496	50 cm depth suggest a faster O_2 intrusion rate compared to the other sensors (Figure 9a), it is not
497	expressed in the E_h recovery time (Figure 9c). During summer all the observed O ₂ curves show
498	steep recovery that ceases after about 20 hours (Figure 9b). According to the O ₂ observations, the
499	gaseous O ₂ intrusion process at 50 and 75 cm depths starts at the very beginning of the drying
500	stage, while at 25 and 100 cm depths there is a two-hour delay (Figure 9b). Nevertheless, the E_h
501	conditions recovery is comparable at all depths (Figure 9d).





- 518 Figure 9: Plot of hourly means of E_h and gaseous O_2 measurements from the beginning of the drying
- 519 stages. The statistics are based on (a,c) 14 cycles during winter (November–April) and (b,d) 19 cycles
- 520 during summer (May–October). Note that the gaseous O_2 (a,b) measurements were obtained at 25, 50, 75
- 521 and 150 cm depths and the E_h conditions (c,d) were observed at 25, 50, 75 and 100 cm depths. All
- 522 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 (400mV <),
- suboxic ($400mV > E_h > 100mV$), and anoxic $E_h < 100mV$) conditions, respectively.

526 **4 Summary and Conclusions**

527 Continuous monitoring of E_h , θ , T, and gaseous O_2 in the vadose zones of SAT infiltration ponds 528 was carried out for about 600 days. SAT operation was subjected to long and short wetting and 529 drying cycles and seasonal changes. The datasets enabled the examination of factors that control 530 the hydrological and geochemical conditions in SAT. Calibrated and validated hydrological 531 models were applied to investigate the water flow dynamics in the SAT vadose zone. E_h and 532 gaseous O_2 observations were averaged to determine seasonal changes and deduce the optimal 533 length of wetting and drying stages.

The examination of the measured E_h conditions illustrated a noticeable decline to markedly 534 negative values (-450 mV >) during summer and to values between 0 and -50 mV during winter. 535 536 These E_h conditions were established following 30 hours of wetting, and no considerable changes in E_h conditions were noticeable until the wetting stages ceased. A monthly statistic of 537 the E_h measurements illustrated the relationship between the size of the E_h amplitude and the 538 seasonal T changes. Furthermore, it is speculated that the limited decrease in E_h conditions 539 540 during winter were due to lower microbial activity. An additional support for this claim is the 541 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_h 542 data were averaged and separated between the winter and summer periods. During the summer 543 period, the optimal time length of a wetting stage, according to E_h observations, is about 30 544 hours. Determining the optimal length of a wetting stage during winter is challenging. 545 Practically, there are no significant differences between E_h conditions during winter and summer 546

for the first 28 hours of wetting. Thus, the length of a wetting stage during winter should be 30hours, as in the summer period.

The length of a drying stage, following the E_h observations, should be about 36 hours, regardless 549 of the initial values of the E_h conditions and the season. Note that during summer, a longer drying 550 551 time is required for the E_h conditions to attain suboxic conditions. Nevertheless, the gaseous O₂ observations indicated faster aeration rates during summer, which compensate for the very 552 negative E_h 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.

Implementing in situ E_h , θ , T and gaseous O_2 sensors provided continuous high-resolution

observations. These datasets revealed the hydrological and biochemical dynamics in SAT as

imposed by seasonal and operational changes. Analysis of the E_h and O_2 measurements enabled

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,

the operational use of a wetting/drying ratio in SAT management should be reconsidered.

563

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