Impact of pre-treatment technologies on soil aquifer treatment

A. Besançon, M. Pidou, P. Jeffrey, B. Jefferson and K. S. Le Corre

ABSTRACT

This study investigates the impact of pre-treatment options on the performances of soil columns simulating soil aquifer treatment (SAT). For this purpose a conventional activated sludge (CAS) process, a membrane bioreactor (MBR) and vertical flow reed beds were used as single units or in combination before SAT. The influent and effluent from each treatment train were monitored over three successive 6-month periods, corresponding to changes in the operational conditions of the MBR and CAS units from 6 days' sludge retention time (SRT) to 12 and 20 days. All the columns acted as efficient polishing steps for solids and bacteria. The column receiving effluent from the CAS system running at 6 days' SRT also presented high total nitrogen and total phosphorus removals, but this column was also associated with the lowest infiltration rates over that period. While the quality of the effluent from the column following the CAS process increased over 18 months of operation, the effluent quality of the columns receiving MBR effluent degraded. No correlations were found between variations in SRT of the MBR and CAS processes and the columns' performances. Overall, all columns, except the one receiving CAS effluent, underwent a reduction in infiltration rate over 18 months.

Key words | conventional activated sludge, membrane bioreactor, reed beds, soil aquifer treatment

A. Besançon
M. Pidou
P. Jeffrey
B. Jefferson
K. S. Le Corre (corresponding author)
Cranfield Water Science Institute,
Cranfield University,
Cranfield,
Bedford,
MK43 0AL,
UK
E-mail: *k.s.lecorrepidou@cranfield.ac.uk*

INTRODUCTION

Soil aquifer treatment (SAT) is used to replenish aquifers with water of impaired quality, hence providing an opportunity to store reclaimed water for future use while reducing the discharge of partially treated effluent to water bodies (Crites *et al.* 2006). In regions suffering from water scarcity and where conditions for ground water recharge using surface infiltration facilities are met, such as land availability, aquifer type and hydrogeology, SAT is increasingly used as either a wastewater-polishing step or a complete treatment system to help augment water resources. Indeed, SAT systems are low-cost, robust and simple to operate while capable of producing water suitable for irrigation and indirect potable reuse (Asano *et al.* 2007).

However, many water reclamation projects for indirect potable reuse have failed due to the perceived health risk associated with the eventual contamination of soils and native groundwater. This led to an increase in the number of schemes, including SAT schemes, using highly treated water sources (e.g. effluents from reverse osmosis (RO) or advanced oxidation processes) for aquifer recharge as a precautionary approach (Lee & Jones-Lee 1996). However, the use of highly treated water for aquifer recharge, and more specifically SAT, may not be economically viable and may also affect the SAT performances. Indeed, as illustrated by Johnson *et al.* (1999), water containing very low carbonates levels such as RO effluents will significantly dissolve minerals, resulting in adverse changes in soil structures. As highlighted by Pavelic *et al.* (2011), the evaluation of treatment options prior to SAT is therefore a major challenge when designing such schemes as a balance needs to be

doi: 10.2166/wrd.2016.163

found between low and high-tech treatment options to limit impacts on infiltration rates, prevent irreversible clogging and groundwater contamination, while remaining economically viable.

Understanding how the level of pre-treatment of effluent can affect SAT performances is therefore crucial. In this context, this study compares the impact of three types of technologies including a conventional activated sludge (CAS) process, a membrane bioreactor (MBR), a vertical flow reed bed (VFRB), used as single units or as multi-barrier systems, on SAT treatment performances and infiltration rates.

MATERIAL AND METHODS

Experimental design

Five pre-treatment trains involving three types of technologies including a CAS process, an MBR, and VFRBs, used as single units or multi-barrier systems, were used prior to soil columns (Figure 1(a)). The MBR and CAS processes were run at an 8 hour hydraulic retention time and successively 6, 20 and 12 days, sludge retention time (SRT). For each SRT, the VFRBs were fed during 6 months.

The soil columns, made of clear PVC tubes, were filled with 10 cm of gravel and 1 m of sieved (2 mm sieve mesh) and repacked aquifer material from the Shafdan SAT site (Israel) at a 1.5 g.cm^{-3} density (Figure 1(b)). The dimensions of the columns were chosen to allow the approximation of the flow rate to one dimension with negligible wall effect. All columns were operated under unsaturated conditions, at rotations of 7 days wetting/7 days drying (7w/7d) cycles and at a constant head of 5 cm to ensure stable hydraulic conditions during the wetting cycles. Running the column at constant head meant that flows through the columns varied over time, ranging from a maximum of 332 mL.d^{-1} for the column fed with MBR effluent (i.e. retention time (RT) of 1.6 days) in the early stages of the experiment, down to a minimum of 3 mL.d^{-1} (i.e. RT of 159 days) for the column receiving effluent from the VFRB in the very last stages of the experiment.

Wastewater analysis

The influent and effluents of the various treatment trains were monitored over 18 months. Over the first 6 months, the CAS and MBR systems were operated at 6 days, SRTs, while over the two following 6-month periods, the SRTs were set at 20 and 12 days, respectively. The results for the VFRB fed with primary effluent are not related to SRT, but refer to three successive periods of 6 months when the CAS and MBR were operated at 6, 20 and 12 d SRT.

All samples were analysed for total suspended solids (TSS) measured according to *Standard Methods* (APHA 1998).

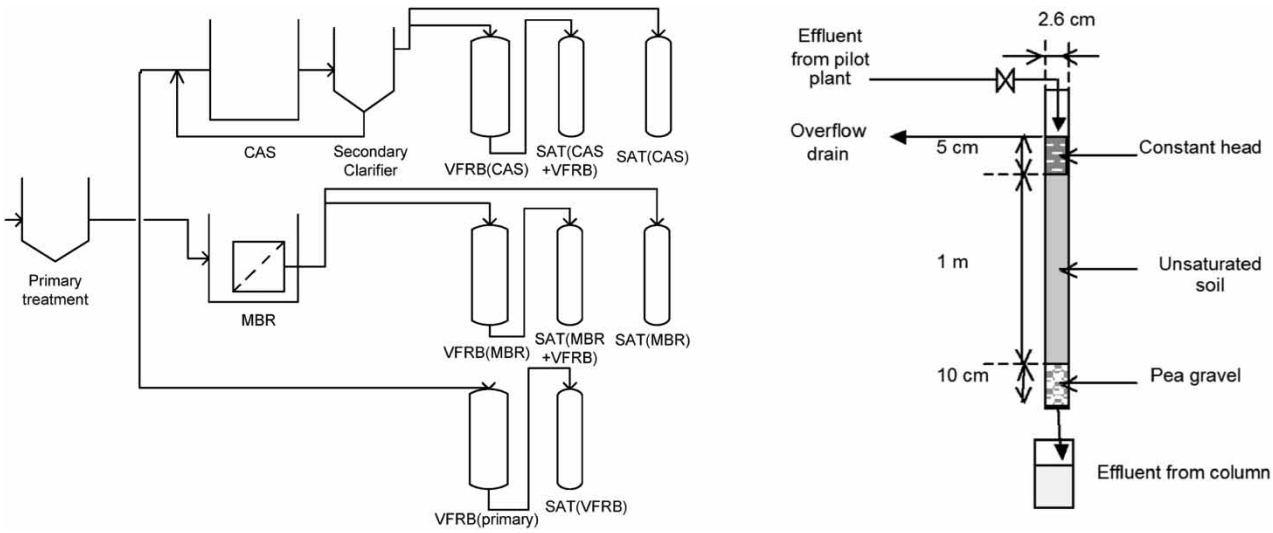

Figure 1 | Scheme of pilot plant treatment trains (a) and soil columns (b).

Conductivity and pH were measured using a combined pH/conductivity meter (Jenway 3540, Bibby Scientific Ltd, Jenway, UK), and turbidity (NTU) was measured using a turbidimeter (HACH 2100N, Camlab Ltd, Cambridge, UK). For each train, influent and effluent chemical oxygen demand (COD), total nitrogen (TN), ammonium (NH_4), nitrates (NO_3) and total phosphorus (TP) were analysed using Merck cell tests (VWR International, UK). In addition, the samples were analysed for total coliforms and *Escherichia coli* (Compact dry EC, HyServe, Germany) as indicators of fecal contamination.

RESULTS AND DISCUSSION

Performance of the treatment trains before soil passage

The characteristics of the influent used (primary effluent) and the performance of each train in removing basic wastewater parameters over each sampling period are summarised in Table 1.

When used as a single treatment, as expected the MBR generally performed better than the CAS and the VFRB over all sampling periods in terms of COD, TSS, turbidity and microbiological contaminants (Table 1). For instance, it achieved 89%, 92% and 91% COD removal at 6, 12 and 20 day SRTs, respectively, as opposed to 79%, 86% and 86% for the CAS systems over the same sampling periods. This suggests retention of particulate or non-biodegradable COD by the MBR. Hasar & Kinaci (2004), who compared CAS and MBR under similar conditions and influent composition, also found the MBR to produce significantly higher effluent quality than CAS. In terms of solids and turbidity, the MBR achieved 99–100% removals at all SRTs with residual values of 1.3 $mg.L^{-1}$ for TSS and 1 NTU for turbidity, while after CAS equivalent values were comprised of between 14.7 and 24.5 $mg.L^{-1}$ for TSS and around 9 NTU for turbidity, corresponding to respective removals of 70–90 and 92–97% (Table 1). These results correlate with other studies producing effluent with less than 2 $mg.L^{-1}$ of TSS and a turbidity of 1 NTU for MBRs (Ng & Hermanowicz 2005) and 14–30 $mg.L^{-1}$ of TSS and a turbidity of 3–5 NTU for CAS (Massé *et al.* 2006). It should be noted that the solids content in the CAS effluent after 6 days' SRT was significantly higher at 25 $mg.L^{-1}$ compared to 15 and 18 $mg.L^{-1}$ at the 12 and 20 day SRTs. This was related to a lower CAS floc strength at 6 days' SRT (data not shown).

For the VFRB used to treat primary effluent, 92–99% TSS and 98–100% turbidity removals were achieved. It should also be noted that the VFRB, when used in combination with the CAS system, achieved high TSS and turbidity removals and further improved the CAS effluent quality by up to 1.9 log in terms of total coliforms and *E. coli* contamination at 6 days' SRT, hence demonstrating the high buffering potential of VFRB for CAS treated effluent. However, as anticipated, VFRB, when fed with MBR effluent, degraded the MBR effluent quality in terms of solids and microbiological contamination (Table 1). In that case, the slight increase in microbiological contaminants in effluent of the VFRB is presumably linked to the presence of coliforms in the VFRB due to cross contamination. Overall, at all SRTs, no *E. coli* were found in the MBR effluent while total coliforms were only observed sporadically (i.e. 0–4 $CFU.mL^{-1}$; >99.99% removal). This can be attributed to the formation of biofilm inside the permeate lines (Zhang & Farahbakhsh 2007). Conversely, *E. coli* and total coliforms were found in the CAS effluent to levels ranging from 240 to 2,068 $CFU.mL^{-1}$ and from 2,080 to 18,624 $CFU.mL^{-1}$, hence resulting in lower removals (i.e. 92.96–99.24%) than those achieved by the MBR (Table 1). These results are comparable to those observed in Zhang & Farahbakhsh's (2007) study.

Finally, the CAS effluent quality was similar to or even higher than the MBR effluent quality in terms of TN, NH_4 and PO_4 concentrations and agreed with other studies under similar conditions (Tchobanoglous & Angelakis 1996; Urbain *et al.* 1998). Higher nitrogen removal with the CAS system is possibly due to the presence of a pre-anoxic zone.

Effect of pre-treatment options on the columns' performances

All columns generally achieved high removal of suspended solids, with undetectable levels of TSS and turbidities below 1 NTU. This was found to be similar to the results reported by Bouwer (1991), who observed more than 90% removal, providing an effluent of less than 1 $mg.L^{-1}$ TSS from an influent of 15 $mg.L^{-1}$.

Table 1 | Average influent quality and removal performances achieved by each pre-treatment train over the three sampling periods

	Sampling period	SRT[d]	COD[a]	TN[a]	NH_4^a	PO_4^a	TSS[a]	Turbidity[b]	Total coliforms[c]	*E. coli*[c]
Influent quality	1	–	236 ± 50	44 ± 5	28.4 ± 2.0	4.3 ± 0.7	81.0 ± 10.0	120 ± 40.1	$2.7.10^5 \pm 1.5.10^5$	$5.7.10^4 \pm 8.9\ 10^3$
	2	–	510 ± 490	52 ± 14	27.8 ± 3.8	11.7 ± 2.9	141.0 ± 49.8	271 ± 119.2	$2.0.10^5 \pm 5.10^2$	$2.4.10^4 \pm 1.0.10^3$
	3	–	329 ± 81	45 ± 3	27.0 ± 3.5	7.8 ± 1.2	145.8 ± 40.0	184 ± 27.9	$1.7.10^5 \pm 2.5.10^4$	$4.0.10^4 \pm 6.5.10^3$
						Removal (%)				
CAS effluent	1	6	79	25	96	0	70	92	92.96	96.32
	2	12	86	46	90	54	90	97	98.45	99.00
	3	20	86	33	97	32	88	95	98.77	99.24
MBR effluent	1	6	89	32	96	7	99	99	99.99	100
	2	12	92	38	96	43	99	100	100	100
	3	20	90	37	97	38	100	100	99.99	100
VFRB effluent	1	–	88	34	98	2	92	98	99.9	99.99
	2	–	93	50	99	57	99	99	99.95	99.96
	3	–	94	44	97	42	94	100	99.87	99.89
CAS + VFRB effluent	1	6	89	39	95	9	94	99	99.91	99.94
	2	12	92	40	95	50	97	99	99.88	99.76
	3	20	93	38	99	41	94	99	99.75	99.84
MBR + VFRB effluent	1	6	80	23	95	5	80	94	95.5	94.97
	2	12	88	29	95	46	94	98	98.67	96.38
	3	20	82	51	96	31	84	96	93.69	91.42

[a]In $mg.L^{-1}$ for influent quality and as % for removals.
[b]In NTU for influent quality and as % for removals.
[c]In $CFU.mL^{-1}$ for influent quality and as log removal unless specified.
[d]In days.

COD concentrations remained between 18 mg.L^{-1} and 26 mg.L^{-1} for all columns' effluents except for the columns receiving effluent from the CAS process, where the COD was higher with values of 38 mg.L^{-1} and 34 mg.L^{-1} at SRTs of 6 and 12 days, respectively. This can be explained by the CAS effluent having a lower fraction of readily biodegradable COD, hence requiring long term RTs to be treated (Fox *et al.* 2001). In terms of removal, the columns achieved 44–60% COD removal when fed with primary effluent treated with VFRB; 8–48% when fed with MBR effluents; 5–40% for the column fed with effluent from the MBR/VFRB train, 23–55% for the column receiving CAS effluent and 13–38% for the SATs column fed with effluent from the CAS/VFRB. The lowest COD removals were usually observed at 6 days' SRT. The results obtained here for the CAS fed column are in the same range as those reported by Ak & Gunduz (2013), who observed COD reductions of about 58% for secondary effluent undergoing SAT using 120 cm long columns with an effective soil depth of 75 cm under 7w/7d cycles.

In terms of organic loading, the results suggest that independently of the pre-treatment used, the higher the loading rate, the higher the mass removed (Figure 2). The lowest average organic load of 0.6 g.m^{-2}.d^{-1} corresponding to a removal rate of 0.08 mg_{COD}.d^{-1} was obtained for the column receiving effluent from the CAS system operated at a 6 days' SRT, while the highest average organic load, 19.1 g.m^{-2}.d^{-1}, corresponding to a removal rate of 5.8 mg_{COD}.d^{-1} was obtained with the column receiving VFRB effluent (Figure 2). For the column receiving effluent from the MBR operated at 6 and 20 days' SRT, average loading rates were among the highest with, respectively, 9.5 g_{COD}.m^{-2}.d^{-1} and 16.6 g_{COD}.m^{-2}.d^{-1}, although the column fed with effluent from the MBR operated at 6 days' SRT resulted in a relatively low COD removal rate of 0.65 mg_{COD}.d^{-1}. Overall, these results showed no evidence of a correlation between the level of pre-treatment used and the performance of the columns at removing organics, suggesting in that case that the choice of one technology over another would have little impact on the performance of SAT.

In terms of nitrogen compounds, the operation of the CAS and MBR systems at shorter SRTs led to lower residual concentrations of TN in the effluent of the columns (Figure 3(a)). For instance, an average residual concentration of 13.1 mg.L^{-1} was obtained for the columns fed with effluent from the CAS system operated at 6 days' SRT compared to an average TN concentration of 31.7 mg.L^{-1} for 12 and 20 days' SRTs. Similarly, the average TN concentration in effluent from the columns fed with effluents from the MBR operated at 6 days' SRT was of 22.4 mg.L^{-1} in comparison to an average TN concentration of 34.8 mg.L^{-1} when operated at 12 and 20 days' SRT. In terms of loading rates, these varied from 0.5 g_{TN}.m^{-2}.d^{-1} for CAS effluent (6 d

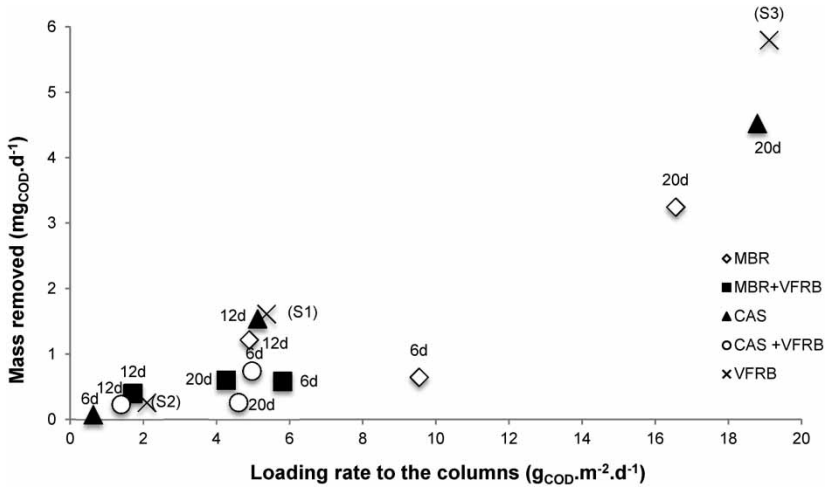

Figure 2 | Average mass removals of organics (as mg_{COD}.d^{-1}) as a function of average loading rates (in g.m^{-2}.d^{-1}) fed to the columns (for the treatment trains involving the CAS and MBR systems, SRTs (days (d)), are indicated on the graph, while for the train involving the VFRB, S1, S2, and S3 indicate the sampling period).

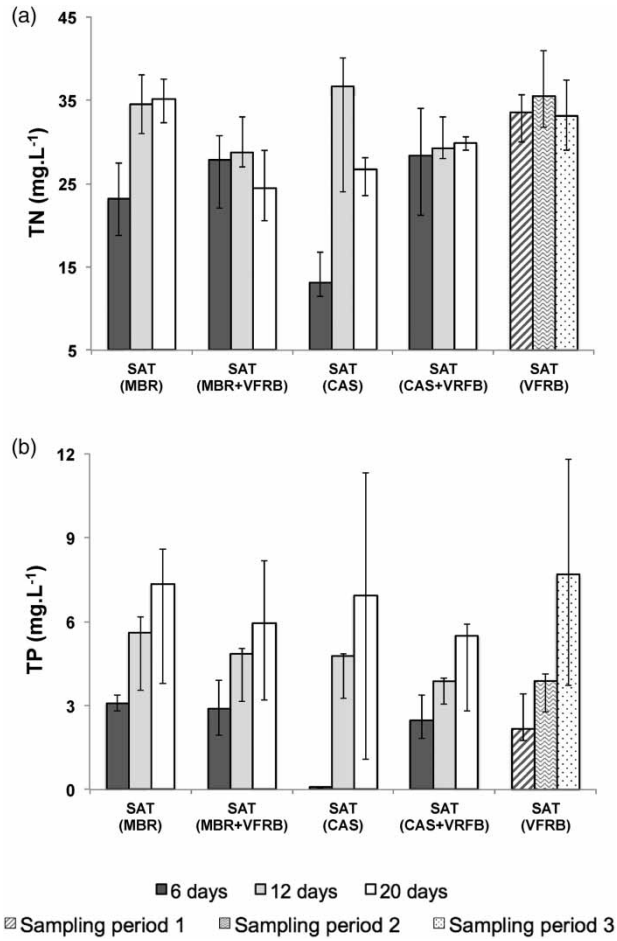

Figure 3 | Total N and P concentrations in the effluents of the columns fed with effluents from the various treatment trains.

SRT) to 17.85 $g_{TN}.m^{-2}.d^{-1}$ for MBR effluent (20 d SRT) corresponding to TN mass removals by the columns of 0.18 $mg_{TN}.d^{-1}$ and 0.60 $mg_{TN}.d^{-1}$, respectively.

Overall, with the exception of the column fed with CAS effluent, where up to 70% TN removal was achieved at a 6 day SRT, the columns generally removed less than 50% of TN. This is in accordance with the literature when conditions for conventional denitrification are absent (Suzuki *et al.* 1992). As conventional denitrification was not expected due to the low carbon to nitrogen ratio of 1.5 in the CAS effluent at 6 days' SRT, the higher TN removal observed after soil passage for this type of effluent may be the result of autotrophic denitrification, such as in Anammox and sulphur-limestone processes (WHO 2004; Fox *et al.* 2006). However, as neither nitrite levels nor levels of sulphur compounds were measured in the present study, it cannot be definitely concluded that these are responsible for the results observed. An alternative explanation is that denitrifying bacteria may have used the carbon source available in the soil (Gable & Fox 2000).

Ammonium removals ranged from 16 to 95% for all processes with lower removals when the pre-treatment processes were operated at 6 days' SRT, especially for the column fed with MBR effluent but usually above 60% for the other columns. The results were generally lower than those observed by Suzuki *et al.* (1992), who found about 90% removal of ammonium when using primary effluent for SAT. A small amount of nitrification was observed in the columns, up to a maximum of 10% for the column receiving effluent from the VFRB fed with primary effluent. This corresponded to the removal of residual ammonia, especially at 6 days' SRT when minimal removal was achieved upstream. On the contrary, at 6 days' SRT no increase in nitrates in the effluent of the columns receiving CAS and MBR was observed. To illustrate, nitrates levels in the effluent of these columns were of 10.8 and 19.8 $mg_N.L^{-1}$ compared to 31.1 and 31.0 $mg_N.L^{-1}$ on average at the other SRTs. Conversely, the nitrate content in the effluent of the columns fed with effluent from the MBR/VFRB treatment train was higher at 12 days' SRT with 28.3 $mg_N.L^{-1}$ compared to 22.7 $mg_N.L^{-1}$ at the other SRTs.

When looking at phosphorus (P) removal after soil passage, these ranged between 36% for the column receiving effluent from the MBR operated at 6 days' SRT to 49% when using effluent from the VFRB fed with primary effluent. However, for all treatment trains involving the MBR and CAS systems operated at 12 and 20 day SRTs, removals below 5% were observed for all columns. The 6 day SRT results are in agreement with those of Kanarek *et al.* (1993) and Lin & Banin (2006), who observed about 50% P removal with long retardation factors and breakthrough times. It should also be noted that over the period where systems were operated at 6 days' SRT, the residual P content in the effluent of the column fed with CAS was 0.09 $mg_P.L^{-1}$ (Figure 3(b)), that is to say, 22–33 times lower than for any other column over that period. This could be linked to the low infiltration rate of 3.5 $cm.d^{-1}$ in that column allowing the slow processes of P mineralisation and sorption to

occur. Indeed in the other columns, infiltration rates were around 20 times higher. In addition, the pH of 8.3 observed in that column, as opposed to 7.3 for others, could enhance or be a result of P precipitation. In general, the pH in the effluents of the columns was 7.8 on average compared to 6.9 in pre-treatment effluents. However, phosphorus leakage was observed after all columns when changing the SRTs from 6 to 20 days. No such trend was measured in influent P content, suggesting a modification or a saturation of the soil sorption capacity.

Finally with regards to indicators of fecal contamination, total coliforms remained below 2 CFU.mL^{-1} in effluent of the columns fed with effluents from the MBR at all SRTs. The use of a CAS treatment, followed or not by VFRB, before soil passage also resulted in low levels of total coliforms in the columns' effluents, respectively below 1 and 5 CFU.mL^{-1} at 12 and 20 days' SRT, and 43 and 108 CFU.mL^{-1} at 6 days' SRT. It should be noted that an increase in total coliforms over time was observed in the columns' effluent for the treatment train combining the MBR and VFRB and the train primarily treating effluent with a VFRB. For instance, over the three sampling periods, total coliforms in the columns effluents increased from 28 to 92 and then 183 CFU.mL^{-1} when using primary effluent treated with a VFRB, and from 1 to 4 and then 13 CFU.mL^{-1} when using the MBR and VFRB as a combined pre-treatment. In addition, the *E. coli* count ranged only between 0 and 3 CFU.mL^{-1} in all columns' effluents.

Coliform removals through soil passage ranged between 54.70 and 99.97%, except when using MBR effluents where the counts were very low. Constant removal above 99.77% at all SRTs was only obtained when using CAS solely as a pre-treatment. This result supports findings already reported in the literature, with more than 99% removals in full-scale SAT systems or longer soil columns treating raw or secondary effluent (Guessab *et al.* 1993; Brissaud *et al.* 1999; Jimenez & Chávez 2004).

Impact of pre-treatment on infiltration rates

With a mean of 34.7 cm.d^{-1}, the infiltration rate in the column fed with MBR effluent was 2.2–2.5 times higher than in the other columns at all times (Figure 4). In addition, this rate was very variable for the first 6 months, ranging between 1.6 and 110 cm.d^{-1}. A significant reduction was observed after 1 year of operation, from an average of 46.3 cm.d^{-1} during the first year to 12.2 cm.d^{-1} for the last 6 months. Similar infiltration rates and decreases over time were observed for columns infiltrated with effluent from the MBR/VFRB and CAS/VFRB treatment trains (Figure 4). A different pattern occurred for the columns fed with single treatment units, namely the VFRB and

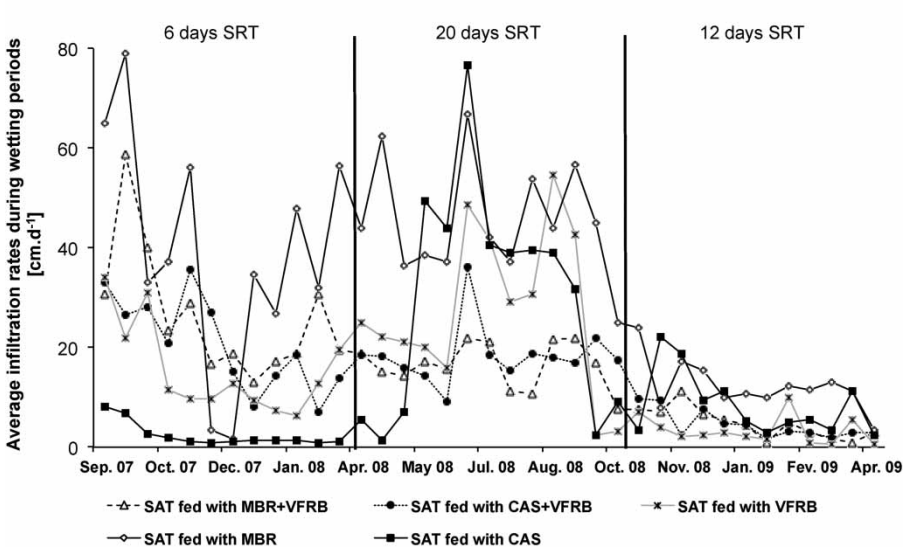

Figure 4 | Infiltration rates in SAT soil fed with treated wastewater.

CAS ones. For instance, the infiltration rate in the column fed with primary effluent treated with a VFRB increased from 16.3 to 30.0 cm.d^{-1} before it decreased to 3.4 cm.d^{-1} in the last period (Figure 4). The column fed with CAS effluent provided the lowest infiltration rate at 6 days' SRT with 2.7 cm.d^{-1} before it increased to 33.7 cm.d^{-1} after 6 months of operation. For the last period, the infiltration rate in that column was higher than over the first period at 8.4 cm.d^{-1}.

Infiltration rates at 12 days' SRT in all columns, after 1 year of operation, and in the column infiltrating effluent from the CAS system operated at a 6 day SRT, were lower than the range observed by Lin *et al.* (2004) at the SAT site from which the soil used in this study originates, where rates varied between 15 and 130 cm.d^{-1}. However, in Lin *et al.*'s (2004) study, the flooding periods were only 1–2 days with 5–7 day drying periods and the pre-treatment, an activated sludge process, included nitrification and denitrification stages. These differences presumably explain these variations in infiltration rates.

The variations in infiltration rates in the columns following CAS solely and VFRB may be related to differences in temperature, which were 7.8 °C, 12.2 °C and 5.4 °C for the 6, 20 and 12 day SRT periods, respectively. Lin *et al.* (2004) also observed an increase of infiltration rate with temperature, and attributed the phenomenon to changes in air and effluent viscosity. The differences in initial infiltration rates between the different columns can be attributed to solids content and nature in the influent. Indeed, the columns fed with MBR effluent received no solids and presented the highest initial infiltration rate, followed in ascending order by the columns fed with effluent from the trains involving VFRBs, and finally the column fed with CAS effluent, which had the highest solids content. It is known that TSS is one of the main physical factors affecting SAT permeability (Pavelic *et al.* 2011).

When using secondary effluent for SAT, physical clogging is usually the major factor affecting infiltration rates, however, in the use of effluent with low SS content over prolonged periods of inundation, as here, biological clogging can become influential, especially sub-surface (Rice 1974). The aerobic activity measured in the soils receiving effluent from the CAS and the VFRB systems (data not shown) were similar to the aerobic activity in other soils despite receiving more nutrients, especially organics. This suggests that the low and variable infiltration rates observed in these two columns may be caused by clogging via facultative anaerobic bacteria (Seki *et al.* 1996; Platzer & Mauch 1997) or extracellular polymeric substances (Vandevivere & Baveye 1992), which are temperature sensitive and controlled.

CONCLUSIONS

When considering treatment performances, the different technology profiles were generally reflected in the columns' performances. The MBR provided a bacteria-free effluent, and the column effluent after this treatment process also had the best bacterial quality. In the same way, the columns fed with VFRB effluents offered constant quality with enhanced nutrient removal. Overall, the soil columns were able to achieve high removals of solids and bacteria after only 1 m of soil passage. The columns fed with effluent from the CAS system set up at 6 days' SRT presented high removal of TN and almost complete removal of TP, but this was also associated with a low infiltration rate, high pH combined with low temperatures. While the quality of the effluent from the column fed with CAS effluent improved over the 18 months of operation, the effluent quality from the column receiving MBR effluent degraded over the same time.

In terms of process operation, no correlations were found between variations in SRT and variations in the performances of the columns. However, it is possible that changes in temperature between the sampling periods affected any such correlations.

The impact of pre-treatment level on infiltration rates was, as expected, closely linked to the ability of the processes at removing solids, with the highest rate obtained when using MBR effluent and the lowest rates when using CAS effluent and effluent from the VFRB treating primary effluents. Overall, all columns, except the one receiving CAS effluent, underwent a reduction in infiltration rate over 18 months. Controversially, it seemed that the lower the infiltration rate, the lower the clogging propensity of the soil but the higher the impact of temperature on the rate.

ACKNOWLEDGEMENTS

This study was performed in the framework of the research project RECLAIM WATER ('Water Reclamation

Technologies for Safe Artificial Groundwater Recharge' supported by the European Commission under Thematic Priority 'Global Change and Ecosystems' of the Sixth Framework Programme (project reference: 18309)). The authors would like to thank Mekorot (Israel) for supplying aquifer material from the Shafdan SAT site for the column experiments.

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First received 30 September 2015; accepted in revised form 8 January 2016. Available online 10 February 2016