CRANFIELD UNIVERSITY

Axelle Besançon

TECHNOLOGIES AND MULTI-BARRIER SYSTEMS FOR SUSTAINABLE GROUNDWATER RECHARGE AND IRRIGATION

SCHOOL OF APPLIED SCIENCES
Department of Sustainable Systems

PhD Thesis
Academic Year: 2006 - 2010

Supervisor: Dr Bruce Jefferson
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This thesis is submitted in partial fulfilment of the requirements for the degree of Doctor of Philosophy
(NB. This section can be removed if the award of the degree is based solely on examination of the thesis)

ABSTRACT

Managed aquifer recharge (MAR) consists of artificially replenishing groundwater to facilitate reuse and/or the associated environmental benefits. Meanwhile, soil aquifer treatment (SAT) is a process of geo-purification designed and operated to improve the quality of the infiltrating water and is thus a type of MAR. SAT consists of a basin operating under rotation of drying and wetting periods. Often, SAT involves water of impaired quality applied onto soil and consequently it implies various risks of health, geochemical and physical nature with difficult or irreversible remediation.

To study the effect of pre-treatment on SAT a pilot plant including conventional activated sludge (CAS), a membrane bioreactor (MBR), tertiary and secondary vertical flow reed beds (VFRB) and SAT soil columns. The sludge retention time (SRT) in the CAS and MBR processes was changed every 6 months to look at the impact of SRT on SAT. Each unit and treatment train effluent was characterised to determine the impact of effluent quality on SAT performance.

This study showed that tertiary VFRB, especially when fed with MBR effluent, was the best option for SAT and irrigation reuse as it provided the best compliance with reuse standards and the best fertilisation potential. However, long-term clogging occurred in SAT after tertiary VFRB, suggesting the need for a longer resting period or shorter wetting period.

This study also highlighted the importance of total suspended solids (TSS) content for SAT removal mechanisms and infiltration rate. In particular, SAT fed with high TSS content effluent was susceptible to temperature variation. Hence the duration of wetting and flooding periods should be adapted according to the season. Further, variation in SRT only indirectly affected pollutants removal by the system including CAS treatment set up at 6 d SRT where the N compounds balance was favourable to an autotrophic N removal.

Keywords: Soil aquifer treatment, MBR, activated sludge, reed bed, sludge retention time, phosphorus, metal.

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“Happiness is nothing more than good health and a bad memory.” *Albert Schweitzer*

“Job’s done.” Saying of a peon (*Warcraft III*)

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ABBREVIATIONS AND NOTATION

AC activated carbon
ANAMMOX anaerobic ammonium oxidation
ANOVA analysis of variance
AOI ioded xenobiotic organic compounds
AOP advanced oxidation process
AOX xenobiotic organic compound
APEC alkylphenol polyethoxycarboxylate
APEO alkylphenol ethoxylates
APHA American Public Health Association
AS activated sludge
ASR aquifer storage and recovery
ASTR aquifer storage transfer and recovery
AWWA American Water Works Association
AWWARF American Water Works Association Research Fondation
b diffusion model constant [mg$_{P}$.g^{-1}]
b_1 Langmuir constant
BNR biological nutrient removal
BOD bio-chemical oxygen demand
BV bed volume
C compliance towards reuse standards [%]
C:N ratio carbon to nitrogen ratio
C_1 floc strength coefficient
CAPEX capital cost
CAS conventional activated sludge
cbz carbamazepine
Ce phosphate concentration at the equilibrium [mg$_{P}$.L-^{1}]
CFU colony-forming unit
C_i initial or feeding phosphate concentration [mg$_{P}$.L-^{1}]
COD chemical oxygen demand [mg$_{O2}$.L^{-1}]
C_t phosphate concentration at the time t [mg$_{P}$.L-^{1}]
$d_{0.5}$ median particle size [µm]
DI dionised (water)
d_{max} maximum floc size [µm]
DOC dissolved organic carbon [mg$_{C}$.L^{-1}]
E. Coli Escherichia Coli
EC European community
EDC endocrine disrupting compound
EEA European Environment Agency
EPA Environmental Protection Agency

EPS extracellular polymeric substances
ESEM environmental scanning electron microscope
F:M food to micro-organisms ratio
FSF free surface flow
G average velocity gradient [s^{-1}]
HFRB horizontal flow reed bed
HRT hydraulic retention time [h]
k adsorption constant [$mg_P.g^{-1}$]
k_0, k_1 and k_2 constants of zero, first and second order kinetics [$mg_P.L^{-1}.h^{-1}$], [h^{-1}] and [$L.mg_P^{-1}.h^{-1}$]
K_1 Temkin constant [$mg_P.g^{-1}$]
K_2 second Temkin constant [$L.mg_P^{-1}$]
K_{YN} Yoon and Nelson rate constant [h^{-1}]
m mass of adsorbent [g]
MBR membrane bioreactor
MCPP 2-(4-chloro-2-methylphenoxy) propanoic acid
MLSS mixed liquor suspended solids [$mg.L^{-1}$]
n modified Freundlich constant
ND non detected
NDMA N-nitrosodymethylamine
NF nano-filtration
NH_4-N ammonium [$mg_N.L^{-1}$]
NO_3-N nitrate [$mg_N.L^{-1}$]
NOM natural organic matter
NRC national research council
NWQMS National Water Quality Management Strategy (Australia)
OM organic matter
OP1EC Octylphenolmonoethoxycarboxylic acid
OPEX operation and maintenance cost
ORP oxido-reduction potential
PAH Polycyclic aromatic hydrocarbon
PO_4-P phosphate [$mg_P.L^{-1}$]
q_e the mass of solute adsorbed per unit mass of adsorbent [$mg_P.g^{-1}$]
q_{max} monolayer capacity or total capacity of adsorbent for adsorbate [$mg_P.g^{-1}$]
r rate constant of diffusion model [$mg_P.h^{0.5}.g^{-1}$]
R robustness ranking
R^2 coefficient of determination
RBC biological rotating contractor
RO reverse osmosis
SAT soil aquifer treatment
SF sand filter
SMP soluble microbial products

SOUR specific oxygen uptake rate [$mg_{O2}.L^{-1}.mg_{Soil}^{-1}.d^{-1}$]
SRB sulphate reducing bacteria
SRT solids retention time [d]
SS suspended solids [$mg.L^{-1}$]
SSF subsurface flow
SSFCW subsurface flow constructed wetland
SUVA specific UV absorbance [$L.mg^{-1}.m^{-1}$]
SVI sludge volume index [$mL.g^{-1}$]
TCP 2,4,6-trichlorophenol
TDS total dissolved solids [$mg.L^{-1}$]
TKN total Kjehldahl nitrogen [$mg.L^{-1}$]
TMP trans-membrane pressure
TN total nitrogen [$mg.L^{-1}$]
TOC total organic carbon [$mg.L^{-1}$]
TP total phosphorous [$mg.L^{-1}$]
TSS total suspended solids [$mg.L^{-1}$]
UF ultra-filtration
UV ultra violet
UV_{254} UV absorbance at the wavelength of 254 nm [m^{-1}]
VFRB vertical flow reed bed
WEF water environment federation
WHO world health organization
x mass of phosphate adsorbed [mg]
α and β Elovich constants [$g.mg_{P}^{-1}$]
γ stable floc size constant
θ breakthrough time when $C_t = 0.5\ C_i$ [h]
σ standard error on regression

1 INTRODUCTION

Project Background

Groundwater is the main source of fresh water in the world and an essential component of the water cycle (Nixon *et al.*, 2000), sustaining soil moisture, geological structure, streams, rivers and wetlands, while providing water for drinking, agricultural and industry (Dillon *et al.*, 2006). To deal with unbalanced extraction and the need for additional water sources, managed aquifer recharge (MAR) has been implemented in both developed and developing countries. It consists of applying or injecting water of impaired quality under controlled conditions. Soil aquifer treatment (SAT) is one of the methods employed in managed aquifer recharge and additionally utilises the soil to further treat the infiltrating water. This additional barrier means wastewater can be applied to SAT. As an important source of reclaimed water, often predictably available irrespective of the climatic variation, wastewater is an important source for MAR (NRC, 1994). The selection of SAT pre-treatment is crucial in reuse planning and usually necessitates extensive preliminary studies including hydrological modelling and soil columns, as well as a continuous assessment of long-term changes in soil and groundwater. Several studies of full scale SAT are available, particularly from Israel and USA, where studies have provided extensive research on SAT soil clogging, performance as well as pre-treatment. Issues and knowledge gaps in SAT include (Bouwer, 1996; Bixio and Wintgens, 2006):

Planning stage

- Adapting policy and local regulations to the SAT project
- Managing the conflict between regulation and optimal SAT pre-treatment
- Managing public acceptance
- Selecting water composition to manage the conflict between preserving soil, groundwater quality and SAT performance

Operational Stage

Monitoring SAT operation and pre-treatment to cope with long-term problems such as:

- Decrease in recharge capacity
- Accumulation of pollutants in soil and risk of release or capacity exhaustion
- Impact of water composition on soil characteristics, including degradation of infiltration rate
- Prevention of pipeline clogging

Project development

The work in this thesis has been initiated for the *Reclaim Water* project with the support of the European Commission under Thematic Priority *Global Change and Ecosystems* of the *Sixth Framework Programme*. The mission of the *Reclaim Water* was to investigate and provide technologies to allow the management of emerging risks from chemicals and pathogens during wastewater reuse for groundwater recharge. The work package including this work focused on water reclamation technology and multi-barriers systems that comply with health and economical constrains in developed and developing countries.

After the wastewater treatment pilot plant, including various technologies and combinations was up and running, SAT soil columns were installed and fed with the pilot plant effluents. After six months of operation and data acquisition after pre-treatment by all units, the operating parameter under investigation was changed in the pre-treatment phase. Most of the parameters monitored were selected according to the preliminary evaluation of the literature and guidelines standards. However, after a few months, distinctive patterns and behaviours in pollutants' removal and soil characteristics were observed. This motivated an in depth study of phosphorus removal and variation in infiltration rate.

Aim and objectives

The purpose of this thesis is to provide a comparative evaluation of technologies options (including multi-barriers systems) for SAT pre-treatment. In particular the thesis considers the appropriateness of using VFRB instead of membrane bioreactors for SAT pre-treatment. Accordingly, the main objectives of this thesis were as follows:

1. Compare different wastewater treatment technologies as potential pre treatments for SAT with particular concern on the impacts of SRT (chapters 3 and 4).
2. Characterise the removal of phosphorus in SAT with reference to the impact of pre-treatment (chapter 5)
3. Understand the role of SRT on the removal of metals in SAT pre-treatment and soil columns (Chapter 6)

Thesis plan

This thesis takes the form of a series of chapters formatted as papers for publication. All papers were written by the first author, Axelle Besançon and have been edited by Dr Bruce Jefferson and Dr Marc Pidou. All experimental work was undertaken by Axelle Besançon with the following exceptions. Chapter 5: electron microscopy has been carried out with the help of Christine Kimpton. Chapters 7 and 8: spectroscopy has been undertaken with the help of Jane Hubble.

Chapter 2, *Treated Wastewater Applied to Soil Aquifer Treatment for Groundwater Recharge: SAT Removals, Soil integrity and Pre-treatment Selection,* gathers literature findings from soil columns to full scale studies to critically assess the current status of knowledge about performance and sustainability of SAT for managed aquifer recharge in order to evaluate relationships between SAT pre-treatment strategies performance of the SAT system itself.

Chapter 3, *Comparison of different wastewater technologies and treatment trains as pre-treatment for soil aquifer treatment and impact of sludge retention time on the systems*, investigates different technologies, and multiple barriers systems, including vertical flow reed bed (VFRB), membrane bio-reactor (MBR) and conventional activated sludge (CAS) in series and as units at 6, 12 and 20 days solids retention time (SRT). The different options of pre-treatment and operation are evaluated for irrigation and SAT reuse in terms of removal mechanism, performance, robustness and compliance with water reuse standards.

Chapter 4, *Impact of pre-treatment trains and their operation on soil aquifer treatment*, focuses on the impact of various pre-treatment technologies and SRT itself on SAT soil and effluent quality.

Chapter 5, *Characterisation of phosphorus removal in soil aquifer treatment and effect of pre-treatment*, aims to understand how various SAT pre-treatments affect phosphate sorption and to elucidate the mechanisms involved. In addition, this paper determines how and to what extent results from short-term studies of soil column and laboratory experiments can be related to long-term experiments and field studies.

In Chapter 6, *Metal removal and the impact of sludge retention time for reuse*, CAS and MBR metal removal mechanisms and performance are compared and the potential benefits of VFRB for SAT pre-treatment in terms of metal removal assessed at different SRTs. Furthermore, this paper looks at the effect of treatment technology and operation on SAT metal removal and its sustainability.

Within Chapter 7, *Discussion: Implications for Water Reuse Projects* the objectives of this study are presented as questions and answered to highlight the scientific contribution to the water reuse knowledge.

Finally, *Conclusions and Future Work*, Chapter 8, lists the key results of the study and provides recommendations on how future investigations can expand current knowledge of SAT and inform pre-treatment selection.

2 TREATED WASTEWATER APPLIED TO SOIL AQUIFER TREATMENT FOR GROUNDWATER RECHARGE: SAT REMOVALS, SOIL INTEGRITY AND PRE-TREATMENT SELECTION

2.1 Introduction

Managed aquifer recharge is a concept of growing interest in a context of increasing ecological awareness and need for sustainable water management in regions facing water scarcity and pollution problems (Hochstrat *et al.*, 2006; Bixio *et al.*, 2006). Currently, one third of the world population is affected by water scarcity and one tenth is suffering from water pollution (WHO, 2009). Climate change and demographic expansion are likely to exacerbate these problems.

Wastewater reuse has a high potential as a source of reclaimed water, because it is available quite independent of seasonal and climatic variation, while its volumes is predictable (NRC, 1994). In Europe, over 200 projects of wastewater reclamation were reported by Bixio *et al.*, (2005), positioning Europe in fourth place just below Japan, USA and Australia. European reuse schemes are essentially agricultural and urban, including groundwater recharge applications. Wastewater effluent following tertiary treatment is the main source for water reuse. Treatment typically includes coagulation and flocculation, filtration and disinfection. However, small-scale constructed wetlands are also quite frequent, particularly in France and Spain, while the Netherlands has larger reclamation facilities with oxidation ditches as secondary treatment (Claasen and Kampf, 2004). Only a small proportion of wastewater reuse projects within Europe is contain soil aquifer treatment (SAT). This contrasts with the situation in the in the Dan region of Israel, where there is a long term experience with the technology of SAT (Idelovitch *et al.*, 2003).

There are different types of managed artificial recharge (Figure 2-1), which can be classified in two categories: subsurface or direct injection and surface spreading, the latter including SAT (De Vries and Simmers, 2002; Lerner, 1990). Major techniques for injection are aquifer storage and recovery (ASR) and aquifer storage transfer and recovery (ASTR), where injection and recovery are from different wells and require a high quality of water to be used. In comparison, surface spreading uses percolation processes to polish or completely treat wastewater and as such less pre-treatment is required. The main configurations adopted for percolation systems include SAT, pond or basin infiltration, surface and sub-surface impoundments and dams in streambeds and wadis.

Numerous studies have revealed the potential of managed aquifer recharge for the removal of various pollutants but also the complex and sometimes irreversible impacts on aquifer soil and water quality (Yaron *et al.*, 2008; Baveye *et al.*, 1998; Bouwer, 1996).

The purpose of this review is to critically assess the current status of knowledge about the performance and sustainability of SAT for managed aquifer recharge in order to evaluate the adequacy of SAT pre-treatments to maintain and enhance SAT system.

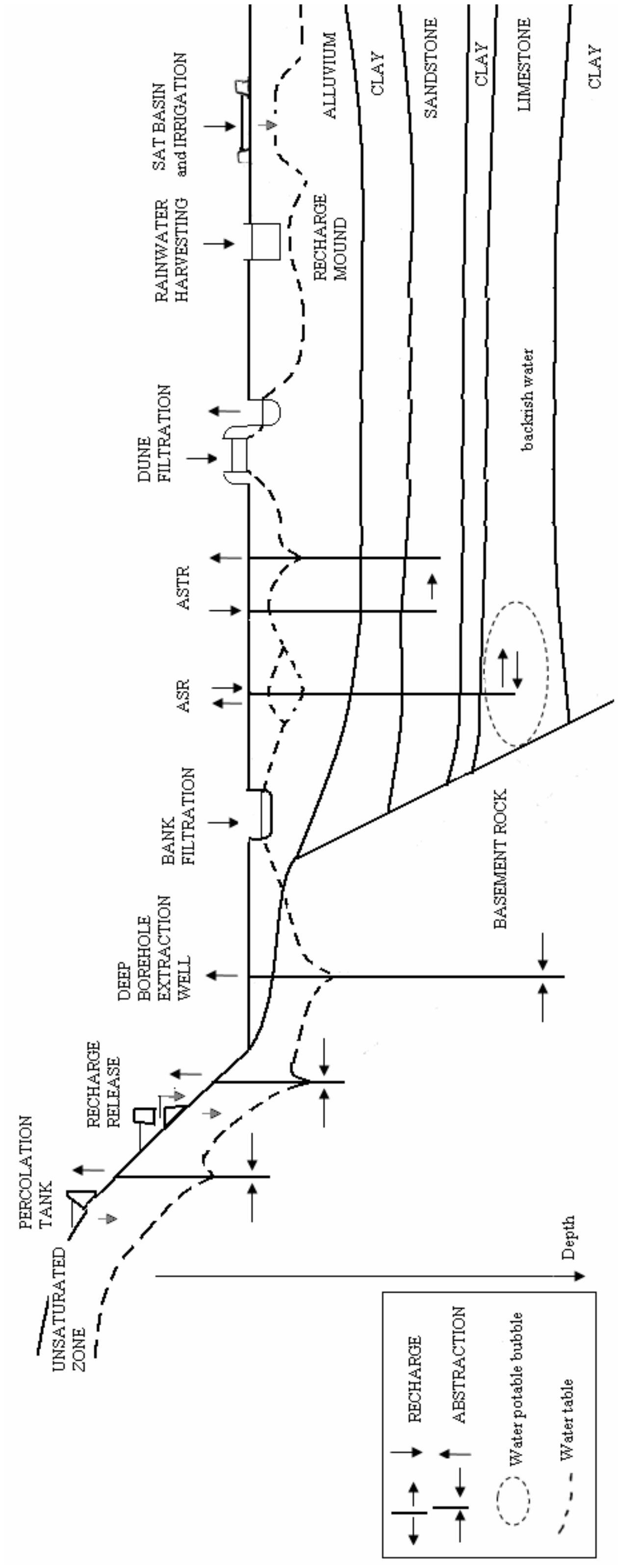


Figure 2-1 Schematic of ground cross-section showing artificial water exchanges with groundwater (Adapted from Amy and Drewes, 2007; Dillon, 2002; EC, 2006).

2.2 Groundwater recharge

2.2.1 Advantages in water reuse for groundwater recharge

Storing water in aquifers presents many advantages over surface water as a potable water source as it provides a more economically, environmentally and socially acceptable option than surface storage (Bouwer, 2002). For instance, the water stored in aquifers is less vulnerable to secondary contamination, by animals or humans, compared to surface water and offers a natural distribution system over large areas (Asano and Cotruvo, 2004; Fox, 1999). Furthermore, managed aquifer recharge provides additional water supplies, reduces saline intrusion and oxygenation of aquifers, while reducing or even reversing the decline of groundwater levels and land subsidence that results from it (Asano and Cotruvo, 2004; Dillon, 2002; Fox, 1999) (Figure 2-2).

The SAT process is essentially a low-cost, simple to operate advanced and robust wastewater treatment (Bouwer, 1991). It also offers an aesthetic advantage over conventional sewage treatment works, since water recovered from SAT is clear, odour-free and comes from a well, drain, or via natural drainage to a surface water source, rather than from a sewer or a treatment plant (EC, 2006). This indirect and natural reuse has been shown to be an important factor in the public acceptance of sewage reuse schemes (Committee on Ground Water Recharge, 1994). For instance, Amy *et al.*, (1993), using secondary effluent, showed that the quality of SAT treated effluent was equal to, or even better than that of a conventional tertiary wastewater treatment plant. Further, costs associated with the SAT option are less than 40 % of the costs of an equivalent in-plant treatment for a similar effluent quality (NRC, 1994). Additionally, compared with direct injection, SAT requires a lower level of wastewater treatment prior to the aquifer recharge, has a longer life cycle and is less expensive (Table 2-1). In fact, the major costs of SAT are the land and distribution system, as capital cost and maintenance of the basin as operational cost, while a well for direct injection has a high capital cost, needs a high level of pre-treatment and flow reversal in the well as maintenance (Fox, 1999).

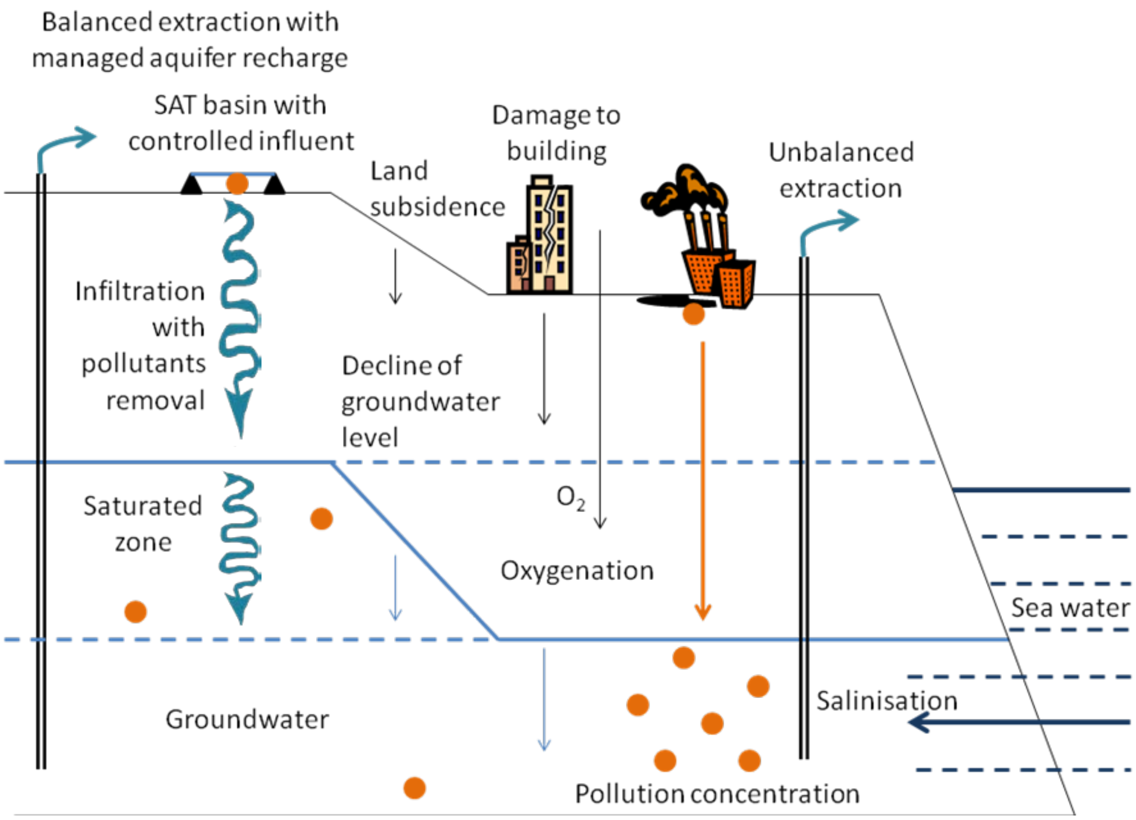


Figure 2-2 Main advantages of managed aquifer recharge (compiled from Candela *et al.*, 2007; Fox, 1999; Levy *et al.*, 2007; NRC, 1998; Sandrin *et al.*, 2001; WHO, 2003; Yolcubal *et al.*, 2003).

Table 2-1 Comparison of SAT and dry well technologies for aquifer recharge (adapted from Amy *et al.*, 1993; Bouwer, 1991*; Fetter, 2001; Fox, 1999; Bixio and Wintgens, 2006^{+}).

	CAPEX	OPEX	Life Cycle (yrs)	Pre-treatment required	Complexity	Reliability	Footprint
Spreading basin (SAT)	land cost <100 k$ US*	0.23-0.25 €.m^{3} $^{+}$	>100	primary to tertiary depending on site	low technology, but advanced treatment	provides an additional barrier to the treatment train	++
Direct Injection (dry well)	500 to 1,500 k$ US*	backflush system	25 to 50	tertiary including disinfection	higher	depends on pre-treatment reliability	--

2.2.2 Issues in managed aquifer recharge

Managed aquifer recharge involving water of impaired quality applied onto soil implies various health, geochemical and physical risks and potentially difficult or irreversible remediation. Many projects of indirect potable reuse have failed on assessment of the health risks, mainly in terms of pathogenic contamination of groundwater (NRC, 1994). Most studies on SAT have been reported to effectively remove bacteria and viruses (Crites, 1985; Brissaud *et al.*, 1999; Jimenez and Chávez, 2004; Guessab *et al.*, 1993; Gold, 1999; Bouwer, 1991; Fox *et al.*, 2006). However, some studies have shown growth and survival of pathogens in aquifer material and faecal contamination of groundwater, with faecal pathogens reaching potable wells (Abbaszadegan *et al.*, 1998). Meanwhile, onsite wastewater treatments and application of wastewater for irrigation have frequently led to faecal contamination of groundwater and disease outbreaks (WHO, 2006). This indicates SAT can on occasion represent an insufficient barrier to pathogenic transport.

Aquifer recharge can also affect the geochemical properties of the soil leading to degradation of groundwater quality and soil clogging. To illustrate, Candela *et al.*, (2007), reported soil dissolution and salinisation of groundwater after irrigation with wastewater. Another underground ecological impact is the development of anaerobic conditions below the recharge basin, changing the position of redox zones and degrading groundwater quality (Greskowiak *et al.*, 2005). The irreversible clogging of subsurface soil is also a major issue in SAT systems, with a wide variety of causes, such as pore blocking by microbial growth and secretion and chemical precipitation (Fox, 1999; Baveye *et al.*, 1998).

Despite these limitations SAT offers efficient removal of wastewater pollutants. The risk of failure to comply with groundwater quality regulations typically arises because of hydro-geological short-circuiting, breakthrough and recalcitrance of certain contaminants (EPA, 2004; NRC, 1998). For instance, pollution of groundwater due to overloading of nitrogen and organic compounds has been reported by Bouwer, (2000). Another risk exists when the infiltrated water from a non-potable aquifer is not confined and subsequently reaches water supply wells (WHO, 2006).

2.2.3 Existing standards and guidelines developed for safe and sustainable aquifer recharge

Numerous guidelines for SAT reuse can be found around the world from international organisations, such as the US- Environmental Protection Agency (EPA), the World Health Organization (WHO) and the European Environment Agency (EEA), to local authorities like the State of California and guidelines values advised in research publications (EPA, 2004; WHO, 2006; EC, 2006) (Table 2-2). The guidelines include definitions of methods for SAT design, operation and monitoring such as setback distance between input and extraction wells and advice on pre-treatment requirements. However, most of the guidelines do not account for SAT removal efficiency, with the exception of Australian guidelines, which encompass water quality levels after SAT passage. The NRC, (1994), based its recharge criteria on the premise that recharged potable groundwater must not require any additional post-treatment to the existing drinking water treatment plant at the point of recovery. In addition, some pollutants of concern and parameters that may impact soil sustainability, like those incorporated in irrigation reuse standards, are not included in these guidelines (Bixio *et al.*, 2005). Nevertheless, the task of setting global standards is difficult because of the complexity of risk assessment with large subsurface volumes and strata, the variability of soil types and the variability of groundwater quality and use (Toze *et al.*, 2009). For these reasons Australian authorities promote local guidelines (NWQMS, 1998).

For indirect potable reuse, a case-by-case approach is recommended, though generally the reclaimed water must have reached drinking water quality before SAT (NWQMS, 1995). For aquifers dedicated to non-potable reuse, the reclaim water quality is related to the intended water use (i.e. irrigation), assuming a confined aquifer. However, such guidelines are not often followed by regulations and enforcement status (Asano, 2007).

Table 2-2 Wastewater reuse standards for SAT and irrigation purposes (Bixio and Wintgens, 2006; EC, 2006; WHO, 2006; WHO, 2003; Kretschmer *et al.*, 2002; Ayers and Westcot, 1985, Pescod, 1992; Asano and Levine, 1998; Rice, 1974).

Parameter	Significance	Polishing goal for SAT	Irrigation unrestricted	Irrigation moderate restriction	Irrigation severe restriction
Total Suspended solids (TSS) [mg.L^{-1}]	TSS can lead to solids deposition on the surface or at depth and consequently anaerobic conditions and soil clogging.	<10	<50	50-100	>100
Organic indicator (COD) [mgO_2.L^{-1}]	Their biodegradation can lead to oxygen depletion. For irrigation excessive amounts can cause problems. Low to moderate concentrations are beneficial for humus and pollutants biodegradation.	70-100	<50	50-100	<100
Coliforms [CFU.mL^{-1}]	*Coliforms* like *E. Coli* are an indicator of microbial and faecal contamination	<100	<10	<100	<100
Nutrients [mg.L^{-1}]	In irrigation they are a beneficial nutrient source for humus and plant growth. However, in excessive amounts they can lead to groundwater contamination and overload sorption capacity in SAT soil.	NH_4-N<2 TN 5-40 NO_3 -N<25	NH_4-N<2 TN<5 NO_3 -N <5 TP<2	TN 5-10 NO_3 -N 5-10	TN 10-30 NO_3 -N 10-30 TP<30
Conductivity [µS.cm^{-1}])	An indicator of dissolved inorganics. Excess of ions like sodium may cause permeability problems in soil.	<2500	<700	700-3000	>3000
pH	The pH affects metal solubility, soil alkalinity and structure and plant growth.	6.5-9.5	6.5-8	6.5-8	6.5-8
Metals [ppb]	Heavy metal can be toxic for humans and the environment and can accumulate in soil and groundwater.	Drinking water	Irrigation reuse		
	Al	100	5000		
	Cr	50	100		
	Co	4	50		
	Cd	3	10		
	Cu	2000	200		
	Mn	400	200		
	Zn	4000	2000		
	Ni	20	200		
	Pb	10	5000		

2.3 Soil Aquifer Treatment

2.3.1 Principle, design and operation

SAT is a process of geo-purification designed and operated to improve the quality of the infiltrating water. SAT consists of a basin operating under a rotation of drying and wetting periods allowing alternating oxic and anoxic conditions to occur. The treatment takes place in three successive zones with contrasting conditions (Figure 2-3). The first metres below the basin correspond to the infiltration zone, with high biological degradation as the influent is nutrient rich. Geochemical interactions such as sorption also occur from this point. The next two zones use percolation processes for treatment and include unsaturated and saturated zones. As the infiltrated water moves down to the aquifer, the unsaturated or vadose zone acts as a natural filter, it also provides disinfection, precipitation and adsorption in association with further biodegradation. From below the water table limit, the saturated zone provides continuous anoxic and anaerobic conditions allowing reduction of nitrates, manganese, iron, sulphates and methane. After the effluent has reached groundwater, during percolation to the extraction well, water continues to be purified with further removal of micro-organisms, precipitation of phosphates and adsorption of synthetic organics (NRC, 1998).

The planning process for SAT is based on land availability, SAT capacity and effectiveness, economical and technical feasibility, criticity of the recharge and quality of both groundwater and reclaimed water (Asano, 1998; Foster *et al.*, 1994). The effectiveness of the SAT itself, and hence the level of pre-treatment required, depends on the soil type, hydrogeology and depth, the operating conditions and the influent quality (Fox *et al.*, 2001; Houston *et al.*, 1999; Quanrud *et al.*, 1996). The design includes the minimum setback distance to extraction wells and the head of water during wetting periods necessary for a proper infiltration rate. This requires consideration of the soil type, the removal level needed, as well as the land availability and water debit to be realised. The design goals are realised through optimisation of the operating conditions, especially the duration of wetting and drying periods. The latter provides the oxic and anoxic conditions, regeneration of the infiltrative zone and of the sorption sites. SAT design and operation are determined using guidelines, pilot plants and hydrological models based on water quality parameters, soil characteristics and basin dimensions (EPA, 2004; Cournoyer and Kriege, 1988; Kool and van Genuchten, 1991).

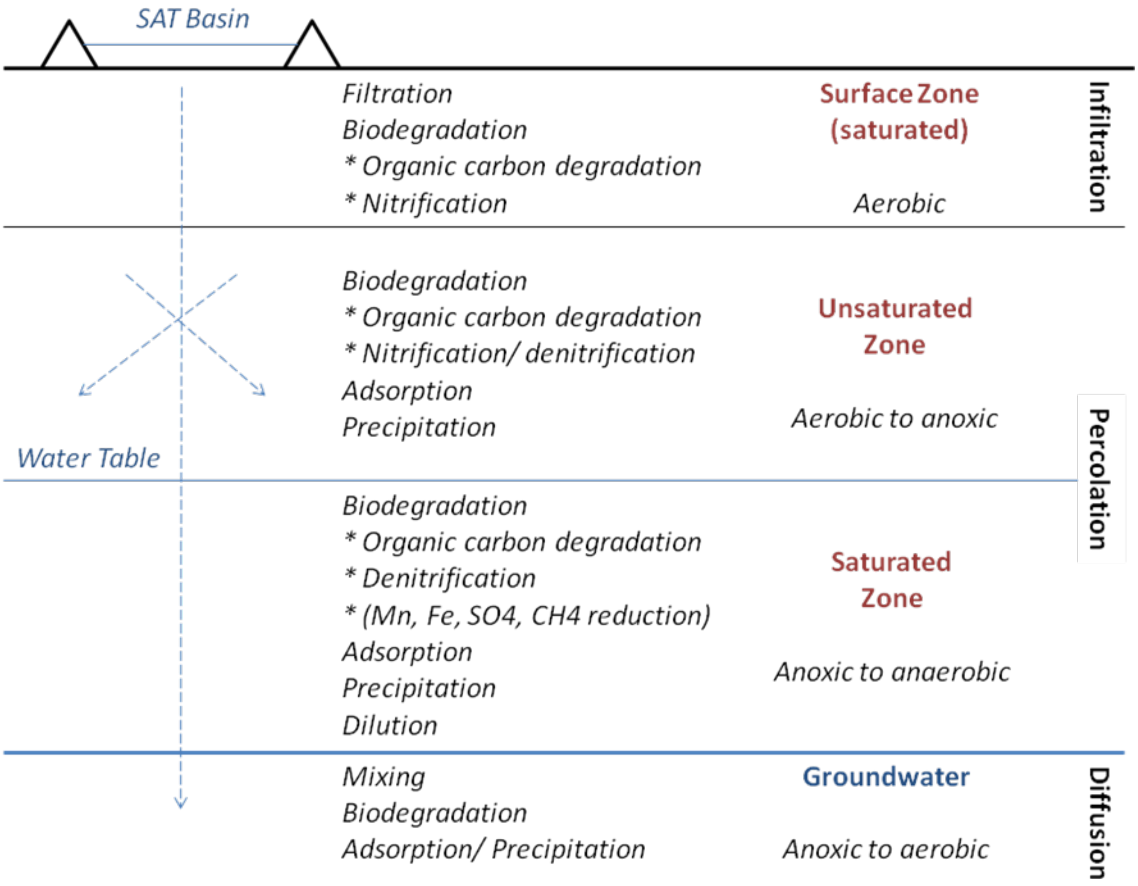


Figure 2-3 Schematic of SAT and its different removal zones.

2.3.2 Pollutants removal efficiency and mechanisms

Organic matter

Anthropogenic organic matter (OM) includes various compounds, such as microbial products and micro-pollutants. It can be classified by characteristics including molecular weight, hydrophobic, humic and fulvic character, biodegradation potential and solubility (Drewes and Fox, 1999). After municipal wastewater treatment, OM is primarily composed of hydrophilic acids at between 40 and 60 % with between 20 and 40 % humic substances and the remainder biodegradable and insoluble fractions (Ma *et al.*, 2001; Imai *et al.*, 2002).

The OM level in recharge waters has been linked to the metal toxicity, the induction of membrane fouling and biological growth, the clogging of SAT soil, and the production of chlorination by-products such as THMs (Cabaniss and Shuman, 1988; Ma *et al.*, 1999; Quanrud *et al.*, 2003; Rauch and Drewes, 2005; Westerhoff and Pinney, 2000).

The mechanisms of OM removal during SAT are mainly based on biodegradation and adsorption, but also include ion exchange, volatilization and chemical reactions (Table 2-3) (Bouwer 1981; Westerhoff and Pinney, 2000). OM is adsorbed onto organic components of soil fine particles by hydrophobic interactions and on minerals by ligand exchange or onto biofilm, especially extracellular products (Carlson, 1995; Fujita *et al.*, 1996; McCarthy *et al.*, 1993). Adsorption of DOC does not depend on either the influent concentration or the presence of acclimated biomass in the soil, but rather the applied readily biodegradable fraction of COD (Fox *et al.*, 2006; Kopchynski *et al.*, 1996; Quanrud *et al.*, 1996). Biodegradation depends on factors such as depth in the vadose zone, oxidation-reduction conditions, nutrients, co-substrates and content of inhibiting substances (Drewes and Jekel, 1996). DOC removal by oxidation-reduction reactions involves oxygen, nitrates and ferric iron (Vanderzalm *et al.*, 2006).

Reported DOC removal by SAT ranges between 20 and 66 % at between 2 and 6 m depth and can exceed 70 % from 15 m depth with effluent content below 3 mg.L^{-1} at this level (Table 2-4) (Fox *et al.*, 2001; Bouwer, 1991; Quanrud *et al.*, 2003). DOC removal is function of short-term removal of easily biodegradable organics and long-term transformation of recalcitrant organics (Fox *et al.*, 2001). For instance, Fox *et al.*, (2006), found that bulk organics were removed primarily by biological treatment, and after 30 d the remaining DOC corresponded mainly to compounds found in drinking water. Amy and Drewes, (2007), also reported significant removal of effluent organic matter, from 66 to 96 % by biodegradation and preferential removal of non-humic compounds. Rauch and Drewes, (2004), observed that 34 and 50 % of the hydrophilic and colloidal organic matter were removed biologically within the upper layer respectively, whereas the hydrophobic fraction and soluble microbial products were recalcitrant in terms of biological or physical removal.

Table 2-3 Mechanisms of organic removal in soil.

Mechanism of removal	Mediated by	Factors affecting removal
ion exchange	minerals and organics in the soil	pH, charge density, molecular weight of DOC
volatilization		pressure, temperature, thermodynamic chemistry of the compound
biodegradation by metabolisation, absorption by cells	micro-organisms, indigenous and applied	depth of the vadose zone, oxidation-reduction conditions, nutrients and co-substrate availability, inhibiting substances, biodegradability of DOC
oxidation-reduction reactions	oxygen, nitrates and ferric iron	oxidation-reduction conditions, concentration of oxidants
adsorption by hydrophobic interactions	organic fraction of the soil	hydrophobicity of bulk organics molecular weight of DOC
adsorption by ligand exchange	mineral fraction of the soil	molecular weight of DOC
adsorption onto biofilm	biofilm that coats the soil particles, especially with extracellular products	hydrophobicity, molecular weight of DOC

Table 2-4 Removal of DOC and COD in SAT.

Reference	soil depth [m]	wetting/ drying [d]	DOC % [mg.L^{-1}]	COD % [mg.L^{-1}]
Amy and Drewes 2007	388	3-10/2-7	76 [1.47]	
	6	3/2	66 [4.84]	
	35		93 [0.98]	
Drewes *et al.* 2003	885		77 [1.23]	
	388		70 [1.56]	
Cha *et al.* 2006	0.5	4/3	48 [2.4]	
	0.5	4/3	35 [3]	
de Feo *et al.* 2007	140-190			67 [32]
Ding *et al.* 1999	9	continuous	[3.9]	
Drewes and Jekel 1996	2	3 d HRT	20 [10-11] (recalcitrant)	
	2	3 w HRT	35	
	2	2 d HRT	23	
	2	5 d HRT	23	
Drewes and Jekel 1998	2	3 d HRT	24.5 [11.4]	
	2	20 d HRT	29.1 [10.7]	
Icekson-Tal *et al.* 2003	15-30	1/2	83 [2]	89 [5.7]
Mottier *et al.* 2000	2			87-91 [35-36]
Viswanathan *et al.* 1999	0.5			70 [7]

Nutrients

Nutrients, including ammonia, nitrate and phosphorus, can have adverse health and environmental effects (WHO, 2006). The removal mechanisms of these nutrients include sorption, volatilisation, transformation by oxidation-reduction processes, precipitation, and biodegradation (Table 2-5) (Mikolalków, 2003; Lee *et al.*, 2006). Ammonium is mainly adsorbed onto soil media particles before soil micro-organisms nitrify ammonium, restoring the initial adsorption capacity (Lee *et al.*, 2006). Anaerobic ammonia oxidation, so called ANAMMOX, has also been reported in Arizona, USA by Gable and Fox, (2000). It is a mechanism of interest in SAT because it is suitable for low carbon to nitrogen (C:N) ratios and when pre-treatment includes only partial nitrification (Fox *et al.*, 2006). The ammonia adsorbed during the wetting period is partly nitrified during the drying period that follows. At the next wetting period, remaining ammonia serves as electron donor for the reduction of nitrates to nitrogen gas, by *Brocadia Anammoxidans* for instance, as follows:

$$NH_4^+ + NO_2^- \xrightarrow{Brocadia\ Anammoxidans} N_{2(g)} + H_2O$$

Drying and infiltration rotations provide aerobic and anoxic alternating conditions, allowing nitrification and conventional denitrification and reducing total nitrogen to less than 10 mg.L^{-1}, with up to 90 % removal of nitrogen present in sewage effluent (Bouwer, 1991). Nitrogen transformation is considered to be mainly a biological process occurring in the first metre of soil, enhanced by the presence of ammonia or other less well-oxidized nitrogen forms (Bouwer, 1996; Mazor, 2004). Conventional nitrogen removal is a function of detention time, C:N ratio and presence of anoxic conditions. The optimum C:N ratio is about 3:1 for anoxic denitrification, with a maximum nitrate content of 10 mgN.L^{-1}. Therefore, when using secondary effluent with nitrification only, a longer flooding period (10 d) is necessary to allow denitrification (Bouwer *et al.*, 1980; Bouwer, 1996; Fox *et al.*, 2006).

Regarding phosphorus removal, adsorption and precipitation are the major pathways. The reactions involve clay, oxides of iron and aluminium, calcium compounds and soil pH (Lin and Banin, 2006). Mineralization and fixing of phosphate by precipitation are slow processes, though sorption sites can be regenerated by wetting and drying cycles. Lin and Banin, (2006), estimated long retardation factors, of between 15 and 235 years and breakthrough times for recovery wells at between 400 and 1100 years.

Numerous studies on SAT have reported effective removal of these pollutants thus producing effluent which met reuse standards (Table 2-6). Up to 100 % ammonium removal has been observed after only 50 cm soil passage (Viswanathan *et al.*, 1999). Ammonium removal forms a gradient in soil depth, with removal from 29 % to leakage at 75 cm and up to 99 % removal after 5.1 m (Ho *et al.*, 1992). Ammonium was found to be preferentially removed under unsaturated conditions with 80 % (Cha *et al.*, 2006). However, the transformation of ammonium led to an increase in nitrates in some studies (Table 2-6). Nevertheless, most research using nitrifying SAT provided nitrate content to below the reuse standard of 25 mgN.L^{-1}, with the exception of the study carried out by Ho *et al.*, (1992), where nitrate content reached 88 mgN.L^{-1}, though only at the lowest depth of 0.75 m.

Denitrification has also occurred in some of the SAT studies with between 32 and 63 % TN removal recorded (Table 2-6). These removals were affected by influent and soil types, with the combination of primary effluent and continuous flooding, or mixed secondary and primary effluents with a drying and wetting cycle giving the highest removal (Ho *et al.,* 1992), and secondary effluent with a drying and wetting cycle giving the lowest removal (Cha *et al.*, 2006).

In terms of phosphorus removal, SAT has proven to be an effective treatment with from 28 to 98 % removal and less than 7 mgP.L^{-1} in effluent despite initial concerns regarding exhaustion of sorption capacity (Table 2-6).

Table 2-5 Mechanisms for inorganic compounds removal in soil.

Mechanism of removal	Mediated by	Factors affecting removal
Adsorption (i.e. ammonia) by ligand exchange and precipitation. Mineralisation (i.e. phosphorus and fluorides)	clays, oxides of iron and aluminium, calcium	pH, oxidation-reduction potential, concentration of oxidants
Volatilization		pressure, temperature, thermodynamic chemistry of the compound
Biological utilisation as cofactor	biomass	depth in the vadose zone, oxidation-reduction conditions, availability of nutrients and co-substrates, inhibiting substances
Adsorption followed by biodegradation: nitrification, denitrification, ANAMMOX	*Nitrifiers*, Denitrifying bacteria and carbon, *Brocasia Anammoxidans*	temperature, loading, retention time, C:N ratio for denitrification, presence of both NH_4 and NO_3 for ANAMMOX, drying/ wetting periods rotation

Table 2-6 Removal of nutrients in SAT.

Reference	soil depth [m]	wetting/ drying [d]	Removal and SAT effluent concentrations % [mg.L^{-1}]			
			TN	NH_4-N	NO_3-N	PO_4-P
Cha *et al.* 2006	0.5	4/3	63 [7]	80 [1.64]	50 [0.88]	58.9 [0.36]
	0.5	Continuous	33 [7]	0 [0.82]	34 [1.32]	97.8 [0.03]
Ding *et al.* 1999	9	Continuous			[1.63]	
Güngör and Ünlü 2005	0.88	7/7			89 [1.2]	
	0.88	7/7			93 [0.9]	
	0.88	7/7			73 [3.3]	
	0.88	3/4			64 [3.3]	
	0.88	3/4			nitrate leakage	
	0.88	3/4			65 [3.2]	
Ho *et al.* 1992	0.75-5.1	9/7	38-46	37 - 80 [13.6 – 4.4]	37 - (-62) [4.2 – 10.9]	28 - 75 [6.7 – 2.3]
	0.75-5.1	Continuous	<0	(-8)- 99 [18.5- 0.2]	<0 [4.9 – 23.4]	64 -31 [2.0 -3.8]
	0.75-5.1	9/12	<0-18	55 - 99 [20 -0.5]	nitrification [88 - 37]	91 - 72 [0.8 -2.6]
	0.75-5.1	9/12	<0-7	88 - 92 [4.6 – 3.2]	nitrification [64 - 33]	97 - 64 [0.3 – 4.0]
	0.75-5.1	Continuous	32-40	29 - 94 [17 – 2.4]	nitrification [12 - 23]	96 - 55 [0.4 – 4.5]
Icekson-Tal *et al.* 2003	15-30	1/2		100 [< 0.02]	nitrification [9.34]	98 [0.05]
Mottier *et al.* 2000	2			98 - 99 [0.3 – 0.5]	nitrification [28.2 – 32.4]	
Viswanathan *et al.* 1999	0.5	7-10/2-3		100 [ND]	21 [3.7]	80 [3.3]

Micro-organisms

Microbial contamination can be attenuated by filtration, adsorption and sedimentation (Table 2-7). The removal efficiency of pathogens, for a given mobility and survival rate, depends on soil characteristics (organic content and surface coating), temperature, residence time, hydraulic conditions, water activity and chemistry (i.e. ionic strength, pH, oxygen concentration) as well as indigenous biomass (Brown *et al.*, 2002; Clement *et al.*, 1997; Jordan *et al.*, 2004; Levy *et al.*, 2007). The characteristics of micro-organisms also determine the predominant mechanism and efficiency of removal. For instance, microbes with appendices will be adsorbed more easily, although low numbers will travel long distance through macropores (Lindqvist and Bengtsson, 1995).

Various experimental conditions have demonstrated almost complete removal of total *Coliforms* (Table 2-8). Jimenez and Chávez, (2004), analysed removals of faecal *Coliforms, Salmonella, E. Histolytica, Shigella* and *Helminth* ova after application of raw wastewater to SAT and found removal percentages between 99.9 and 100 %. However, Levy *et al.*, (2007), found significant differences in *E.Coli* removal depending on sediment characteristics. Removal ranged from 41 % using sediment core with a median grain size of 5 mm to more than 99 % with a grain size of 1 mm. Although chlorination prior to SAT reduces bacterial concentration, it is not recommended as the chlorinated organic compounds formed, such as trihalomethanes, represent a greater risk to the groundwater safety than the potential transmission of a few bacteria or viruses (Singer, 1999). Moreover, residual chlorine could destroy the native biomass of the soil, which contributes to biodegradation. Bond endotoxins are removed mainly by filtration, with up to 90 % removal of total endotoxins from percolation when secondary effluent is applied to soil. Nevertheless rainfall may enhance their transport, mainly for the free endotoxins (Goyal *et al.*, 1980).

Table 2-7 Summary of mechanisms and implied factors for micro-organisms removal.

Mechanism of removal	Substrate	Factors affecting removal
filtration	soil particles	microbial size, residence time, hydraulic conditions, soil particle characteristics (aggregation, porosity)
adsorption	soil particles aggregation	presence of appendices, soil characteristics (organic content and coating)
sedimentation	soil	depth, microbial molecular weight
natural selection/ competition and predators	indigenous biomass	species (intrinsic mobility and survival rate), temperature, residence time, hydraulic conditions, water activity and chemistry (ionic strength, pH, DO)

Table 2-8 Total *Coliforms* removal in SAT.

Reference	soil depth [m]	wetting/ drying [d]	total *Coliforms* % [CFU.100mL^{-1}]
de Feo *et al.* 2007	140-190		98.7 [5 x10^{5}]
Ho *et al.* 1992	0.75-15.1	9/7	90.5 – 99.2 [4x10^{3} - 290]
	0.75-15.1	continuous	85.8 -99.9 [2x10^{4} – 1]
	0.75-15.1	9/12	66 – 99.9 [4x10^{4} – 10]
	0.75-15.1	9/12	99.7 – 99.9 [10^{3} - 1]
	0.75-15.1	continuous	99.9 [2x10^{3} - 47]
Icekson-Tal *et al.* 2003	15-30	1/2	100 [ND]
Jimenez and Chávez 2004	140-190		99.9 [0-330]
Mottier *et al.* 2000	2		78
Viswanathan *et al.* 1999	0.5		99 [80] (Faecal *Coliforms*)
Al-Otaibi *et al.* 2001			100 [ND]

Viruses

Viruses are primarily removed by adsorption to the soil surface (Lance *et al.*, 1976) (Table 2-9). The adsorption of viruses within SAT is based on electrostatic and chemical interactions. Factors affecting their survival and transport are climatic (i.e. temperature, hydraulic conditions, rainfall elution), soil adsorption capacity (i.e. related to pH, dissolved salts, organic matter), aggregation of viruses. Finally low microbiological activity reduces predation and competition (WHO, 2003; Wellings *et al.*, 1975). High concentrations of metal oxides, and divalent cations, low pH and hydrophobicity promote adsorption, while surface-active substances enhance mobility through soil by competing for adsorption sites. Examples of such species are phosphate and DOC (Blanford *et al.*, 2005; Chu *et al.*, 2001; Jin *et al.*, 2000; Powelson *et al.*, 1993; Quanrud *et al.*, 2003).

Powelson *et al.*, (1993), studied removal of the bacteriophages PRD-1 and MS2 and observed between 37 and 99.7 % removal of viruses at 4.3 m depth. This range is due to variation in applied effluent types, which affected PRD-1 removal but not MS2. Blanc and Nasser, (1996), also observed a lower adsorption of MS2 compared to PRD-1 at 14.9-91.6 % and 92.7-99.2 % respectively in loamy and sandy soil, though found considered the effect of water composition insignificant. Fox *et al.*, (2006), suggested that more than 99.99 % removal of bacteriophages could be expected after 30 m and Bouwer, (1991), reported 100 % removal with an influent containing 21 viruses.L^{-1} (Table 2-10).

Table 2-9 Summary of mechanisms and implied factors for virus removal.

Mechanisms of removal	Mediated by	Factors affecting removal
adsorption based on electrostatic and hydrophobic interactions	soil particles	sorption capacity of soil, infiltration rate, temperature, virus type, rainfall dilution, presence of metal oxides and divalent cations, pH, surface active substances, soil saturation, concentration of anions (phosphates) and dissolved organics
predation	microbial predators	Temperature, pH, redox potential, microbial ecosystem

Table 2-10 Virus removal and die-off (adapted from Blanc and Nasser, 1996).

Virus	Conditions	Removal [%]	die off [Log10]
	adsorption on loamy soil		20 d incubation, at 10°C/23°C
MS2 bacteriophage	groundwater	14.9	-1.1/-3.2
	secondary effluent	15.5	-0.9/-4.6
Poliovirus 1	groundwater	99.5	-0.3/-1.4
	secondary effluent	99.3	-0.3/-1.5
PRD-1	groundwater	82.7	-0.6/-1.1
	secondary effluent	93.8	-0.7/-1.1
	adsorption on sandy soil		
MS2 bacteriophage	groundwater	91.6	-2.4/-3.8
	secondary effluent	76.6	-1.2/-5.0
Poliovirus 1	groundwater	77.9	-0.3/-0.5
	secondary effluent	27.3	-0.2/-1.5
PRD-1	groundwater	97.4	-0.5/-0.7
	secondary effluent	99.2	-0.8/-1.2

Solids and colloids

Suspended solids are attenuated by filtration and sedimentation, with more than 90 % removal typical, providing an effluent of less than 1 mg.L^{-1} total suspended solids content (TSS) (Bouwer, 1991; Icekson-Tal *et al.*, 2003; Jimenez and Chávez, 2004). TSS may clog the surface layer of the SAT basin, though appropriate drying periods with surface scraping restores most of the permeability lost this way.

Bradford and Bettahar, (2006), studied negatively charged colloids transport through saturated porous media using columns with different colloid concentrations at sizes of 1.0 and 3.2 µm, as well as sand grain size (Table 2-11). According to their conclusions, colloid retention is enhanced by finer sand grains and higher colloid size and occurs primarily in the upper layers. However, they observed reduced retention when influent colloid concentration increased. Regarding carboxylated hydrophobic colloids, they are retained less because of their higher electrophoretic mobility, while their retention is also less density dependant than other colloid types (Lindqvist and Bengtsson, 1995). The same authors found that charge interactions in addition to hydrophobic interactions were a significant factor for sorption in soil impoverished in organic carbon. The behaviour of polyanionic colloids can be comparable to bacteria (Lindqvist and Bengtsson, 1995; Peterson, 1947). In that case, sorption is principally driven by pH, though also influenced by ionic strength (especially divalent ions) (Goldschmid *et al.*, 1972; Scholl *et al.*, 1990).

Table 2-11 Experimental set-ups for colloids transport studies.

colloid size [µm]	column sand	colloid type and concentration	reference
0.51	630 µm screened (with over 95 % in the 25-630 µm fraction)	carboxylated, hydrophobic, 1.9E+7 spheres.L^{-1}	Lindqvist and Bengtsson, (1995)
1	360, 240 and 150 µm	latex, negatively charged, 7.72E+10, 3.86E+10, 1.93E+10 and 9.65E+9 N.L^{-1}	Bradford and Bettahar, (2006)
3.2	360, 240 and 150 µm	latex, negatively charged, 2.36E+9, 1.18E+9, 5.90E+8 and 2.95E+8 N.L^{-1}	

Micro-pollutants

Of the various anthropogenic micro-pollutants found in water, adsorbable organic halogen (AOX), such as trihalomethanes (THMs), can be biodegraded, volatilized or adsorbed. However, the sorption capacity of soil has little impact on dehalogenated compounds (Bouwer 1981). Drewes and Jekel, (1996), found reductions of DOC and AOX of 23 % and 16 % respectively under aerobic conditions in soil by thiolytical, hydrolytical and oxygenated dehalogenation, while Muller *et al.*, (1988), reported equivalent values of 35 % and 22 % under anaerobic conditions by reductive dehalogenation. Molecular studies of microcosm and chlorinated ethenes have confirmed the existence of natural biological attenuation in anaerobic aquifers (Nijenhuis *et al.*, 2007). However, products from such degradation may be more toxic than the original compound. For example, vinyl chloride may be produced by degradation of ethane chloride under anaerobic conditions. Fox *et al.*, (2006), found THM formation potential (THMFP) between 50 to 65 $\mu gTHMFP.mgDOC^{-1}$ from chlorination, while significant AOX content, mainly AOI (iodinated AOX), persisted for more than 18 months.

Pharmaceuticals, nonylphenol ethoxylate metabolites and other anthropogenic contaminants are an environmental issue, due to their endocrine-disrupting properties (Snyder *et al.*, 2009). Indeed, endocrine disrupting compounds (EDCs) can exert adverse physiological effects on humans and wildlife populations in terms of fertility and sexual development. While applied to SAT, EDCs are easily adsorbed onto porous media, with typical removal above 80 %, and can also be biodegraded under aerobic and biotic conditions (Mansell *et al.*, 2004) (Table 2-12 and 13). Lipid regulators and analgesics have been reported to be removed by SAT, however antiepileptic drugs and mezapine persist (Drewes *et al.*, 2002; Fox *et al.*, 2006). To illustrate, 17β-estradiol, which is the endogenous molecule mimicked by more than 50 non-steroidal anthropogenic chemicals, is removed by more than 99 %, resulting in an effluent content below 1 $ng.L^{-1}$ (Fox *et al.*, 2006). Conroy *et al.*, (2005), also reported also substantial removals of oestrogen antagonists though breakthroughs of oestrogen agonists.

Surfactants, such as alkylphenol polyethoxylates (APEOs), are attenuated within the first few metres of subsurface when alternating flooding and drying periods are applied, so that recovered water contains only low concentrations of alkylphenoxy acetic acids, carboxyalkylphenoxy acetic acids and alkylphenols (Table 2-12 and 13) (Fox *et al.*, 2006; Montgomery-Brown *et al.*, 2003). Fox *et al.*, (2001), also observed high removals of other trace anthropogenic compounds such as alkylphenol polyethoxycarboxylates (APECs), naphthalene dicarboxylic acid, absorbable organic chlorine and absorbable organic bromine. According to Wild and Reinhard (1999), OP1EC, an APEC model compound, is co-metabolized with DOC and reach refractory levels after less than one hour residence time in soil column experiments. Polycyclic aromatic hydrocarbons (PAHs) are retained by adsorption onto soil organic or mineral matter, particularly by small organic soil particles and metal oxides and when the organic carbon fraction of the soil is low. However, with certain types of soil, this adsorption is limited by facilitated transport in the presence of aromatic free NOM and colloids in the mobile phase (Chi and Amy, 2004; Spark and Swift, 2002). N-nitrosodymethylamine (NDMA), a recognized toxic and anticipated human carcinogen pollutant, is naturally attenuated by mineralization mediated by soil micro-organisms under either aerobic or anoxic conditions (Fox *et al.*, 2006).

Rav-Acha *et al.*, (1996), studied breakthrough and adsorption of some micro-pollutants including indole, 2,4,6-trichlorophenol (TCP), naphthalene and fluoranthen. As demonstrated for colloids, they found that hydrophobic compounds were more subject to dispersion and diffusion through the soil material, and noticed that indole and fluoranthen had a slower adsorption-desorption kinetics, with pH dependency.

Pesticides have been recovered through extraction wells and hence may be of concern in some areas (Tuxen *et al.*, 2000). However, high degradation rates of MCPP (2-(4-chloro-2-methylphenoxy) propanoic acid) have been found in aerobic SAT (Tuxen *et al.*, 2000). The herbicide sulfonylurea and its metabolites can be highly mobile depending on the pH and the soil characteristics such as concentration of organic matter and nature of solid surface. However, in general degradation occurs rapidly except for one or two metabolites that persist (Martins and Mermoud, 1999).

Table 2-12 Summary of mechanisms and implied factors for specific substances, trace organic and anthropogenic compounds removal.

Substances	Mechanism of removal	Mediated by	Factors affecting removal
AOX (i.e. THMs)	adsorption, volatilization, biodegradation	soil particles, biomass	oxidation-reduction potential
EDCs (steroids/ oestradiol)	adsorption, biodegradation	soil particles, biomass	soil porosity, DO
Surfactants (APEOs, APECs, NDMA)	adsorption, biodegradation/ mineralisation	soil particles (organics and minerals), biomass,	DOC (co-metabolisation), absence of aromatic free NOM and colloids, substance hydrophobicity, pH
Pesticides/ herbicides	adsorption, biodegradation	soil particles (organics), biomass	pH, soil type (organic content, surface)

Table 2-13 Removals of some micro-pollutants.

Compound	Removal [%]	Reference
APECs CAPECs Octylphenol and nonylphenol	95.5 99.7 80	Montgomery-Brown *et al.*, (2003)
OP1EC, feed [µg.L-1]: 2000 1000 50	 99.9 99.8 99.6	Wild and Reinhard, (1999)
Antiepileptic drug	<0 % 610 ng.L^{-1} Carbamazepine; 155 ng.L^{-1} Primidone	Drewes *et al.,* (2002)
Anti-inflammatory drugs	<99 % except Propyphenazone with 25 %	
Lipid-regulating drugs	100	
Detergents	>64	Icekson-Tal *et al.,* (2003)
Phenol	>83	
Antiepileptic drug (Carbamazepine)	<30	Kreuzinger *et al., (*2004)
Lipid-regulating drugs (Bezafibrate)	no significant removal but 99 % removal in the wastewater treatment plant	

The presence of metals is of concern in water reuse as it has been identified as toxic and EDCs for the aquatic environment (Lester, 1987; Brikett and Lester, 2003). Removal mechanisms for metals during SAT involve adsorption, precipitation, biotransformation and immobilisation into soil, with retention depending on the individual metal and the biological and chemical environment (Alloway, 1995; Vanbroekhoven *et al.*, 2007) (Table 2-14). Heavy metals are mainly retained by complexation onto iron oxides and carbonate, depending on the soil pH as well as surface precipitation within the first metre of the SAT soil (Lin *et al.*, 2004). The main factors affecting their removal are the sorption capacity of soil, pH, oxidation-reduction potential of the microbial community, concentration of heavy metals infiltrated, uptake by plants, TOC, and to a lower extent, particle size and flow rate (Kirkham, 2006; Lee *et al.*, 2004). Adsorbent types such as hydroxides, clays, sulphides, natural organic carbon (NOM) and ions also affect immobilisation and transformation of metals (Wang and Mulligan, 2006). The pH is an important factor for the mobility of metals in soil, however Lee *et al.*, (2004), did not observe desorption by imitating acid rain on SAT. Drewes and Jekel, (1996), also noticed that municipal wastewater reuse provided a consistent neutral soil pH value that did not re-dissolve precipitated metals in soil.

To illustrate the mechanisms of heavy metal removal, arsenic can be adsorbed onto hydroxides, mainly iron oxides, and mineral clays, transformed by oxidation-reduction reactions with sulphides and NOM or bio-transformed by reduction, oxidation, precipitation, methylation and demethylation (Wang and Mulligan, 2006).

A wide range in metal removal efficiency has been reported for SAT as the removal depends highly on the soil type (Kirkham, 2006) (Table 2-15). De Feo *et al.*, (2007), and Icekson *et al.*, (2003), observed low metal retention but with very low influent metal content. However, they also found leakage of Al, Fe and Mn. Similarly, Lin *et al.*, (2004), found that the oxidation of organics during SAT caused reductive dissolution of Mn oxides leading to leakage. Conversely, other authors found metal removal between 67 and 98 % removal independent of the metal in a variety of soil types (Jimenez and Chávez, 2004; Aziz *et al.,* 2008).

Table 2-14 Summary of mechanisms and implied factors for metals removal.

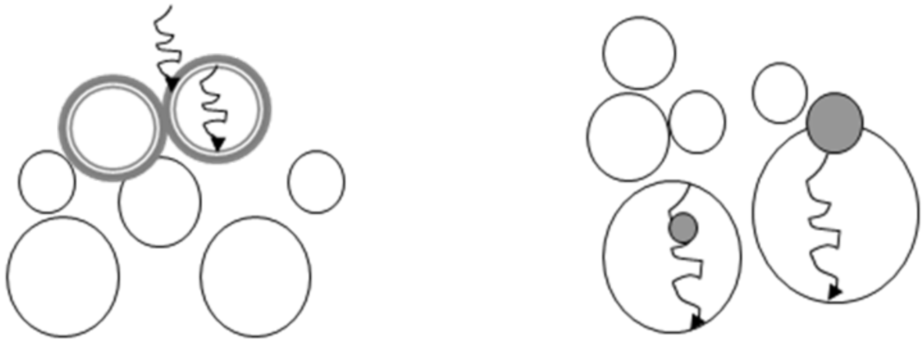
Mechanism of removal	**Mediated by**	**Factors affecting removal**
adsorption/ immobilisation	clays, ions, iron oxides, carbonates	pH, oxidation-reduction potential, concentration of oxidants, sorption capacity of the soil, metal loading, TOC, particle size, flow rate
precipitation/ oxidation-reduction reaction	hydroxides, sulphides, NOM	pH, oxidation-reduction potential
biotransformation: methylation/ demethylation	biomass	depth in the vadose zone, oxidation-reduction potential, nutrients and co-substrates availability, absence of inhibiting substances

Table 2-15 Removal of metals in SAT.

Reference	**Metal removal and SAT effluent content % [ppb]**							
	Al	**Pb**	**Cr**	**Cu**	**Fe**	**Mn**	**Co**	**Ni**
de Feo *et al.* 2007	-19 [57.1]	87 [0.7]	48 [0.3]	30 [3.3]	-25 [110]			
Icekson *et al.* 2003		0 [<3]	25 [3]	0 [<3]	83 [15]	<0 [409]	0 [<3]	45 [6]
Jimenez and Chávez 2004	96-98 [0-140]	78-84 [0-80]	90-91 [0- 2000]	67-82 [0-70]	86-96 [0-940]	88-95 [0-60]		
Aziz *et al.* 2008		93-95	82-91 (CrIII)	96-98				94-98

2.3.3 Irreversible clogging of SAT

Deep clogging phenomena also known as long-term or irreversible clogging occur in depth in SAT basins. Compared with surface clogging of SAT basins, easily removed during the drying period, as implied by the name deep clogging can be irreversible. The main concern is that deep clogging affects infiltration rate over long-term and may require larger land areas to compensate for it. The life cycle of the vadose zone is difficult to estimate, but once clogged, consequences are irreversible since there is no possibility of backwashing or drying such as can occur in direct injection wells or the surface of a SAT basin (Fox, 1999). There is a wide variety of clogging behaviours and mechanisms reported in the literature (Figure 2-4). The phenomenon is usually a combination of surface, physical, chemical and biological clogging mechanisms, and depends highly on the soil aquifer properties, SAT operation, design and applied wastewater composition.



Coating (biofilm, extracellular product, chemical precipitate…)

Pore obstruction (by micro-colonies, clays, precipitates…)

Dispersion of soil particles (from soil composition destabilization)

Compaction (from desaturation, soil destabilization, compression)

Figure 2-4 Scheme of main clogging mechanisms.

Physical Clogging

Clogging by particulate matter occurs mainly at the basin surface and the clogging layer is usually composed of solids, algae, dust, microbes and salt precipitates (AWWARF, 1998). In such cases, amelioration is achieved by scraping or removing the clogging layer during the drying period with good recovery of permeability (Bouwer, 2002). Physical clogging is probably the easiest mode of clogging to control. Nevertheless, in some cases, infiltration of secondary sewage effluent showed that physical clogging is more important than biological in reducing infiltration during short periods of flooding (Rice, 1974).

The concentration of suspended solids, and especially the non-biodegradable fraction, is one of the main physical factors affecting SAT permeability (Loffler, 1969; Okubo and Matsumoto, 1983; Siegrist and Boyle, 1987) (Figure 2-5). Consequently, in order to optimize infiltration, it is recommended to keep suspended solids below 10 mg.L^{-1} (Rice, 1974). This is also illustrated by Figure 2-5, where after 10 days' operation, specific discharge was 3.7 m.d^{-1} at a TSS of 1.4 mg.L^{-1} compared with 0.3 m.d^{-1} at 14.6 mg.L^{-1} TSS. Nevertheless, experiments have showed clogging does not depend primarily on porosity reduction indicating solids concentration is not always the critical factor (Mays and Hunt, 2005). For instance, entrapped gas in soil from desaturation increases gas pressure and therefore saturation, weakening the cohesive forces between particles, so that dispersed matter clogs pores (Faibishenko, 1984; Ferh *et al.*, 1992; Goldenberg *et al.*, 1993; Mazor, 2004; Ragusa *et al.*, 1994; Seki *et al.*, 1998; Allison, 1947). Another example of physical clogging unrelated to particle deposition is land subsidence and compaction. They can be caused by the head of water in the recharge basin, a decrease of the water table or dissolution of soil media (Faibishenko, 1984; Fox, 1999).

The physical properties of soil, notably grain size distribution, influences clogging through pore size, hydraulic capacity and recovery from clogging. Generally, the finer the grain, the lower the water transfer and the higher the permeability recovery (Table 2-16) (Nevo and Mitchell, 1967; Platzer and

Mauch, 1997). For a given mass of clogging particles, experimentations on saturated porous media showed that greater fluid velocity leads to greater permeability (Kau and Lawler, 1995; Narayan *et al.*, 1997; Veerapaneni and Wiesner, 1997). In other studies, smaller fluid velocities have greater head loss, when the ratio of maximum to minimum velocity remains below 3.5 (Mays and Hunt, 2005).

Table 2-16 Physical causes and factors reducing soil permeability.

Factor reducing permeability of soil	Cause
Land subsidence and compaction	High head of water in the recharge basin, decrease of the water table
Physical properties of soil: particle size distribution; particle shape and texture; voids ratio; pore size ; saturation; hydraulic capacity; capacity to recover from clogging	Finer grain (lower initial permeability) lowers the water transfer but increases recovery
Pore obstruction but not primarily porosity reduction.	Solids and organic matter clogging pore spaces or cells sieving at pore throats.
Increase of pressure and desaturation of soil	Entrapped gas in soil, weakening cohesive forces between particles, so that dispersed matter clogs pores
Low fluid velocity (for a given mass of clogging particles, with saturated porous media)	High head loss, increase depth of particle penetration and deposit morphology

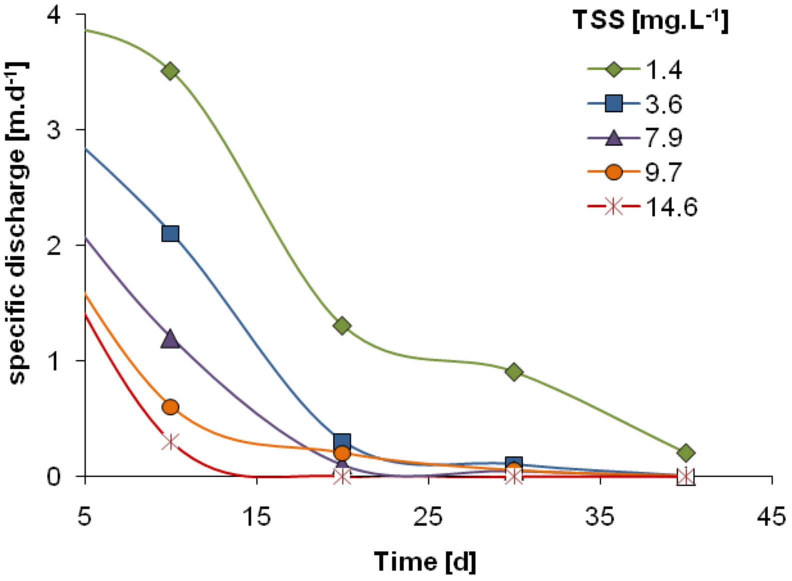


Figure 2-5 Decrease in specific discharge with time at different TSS inputs. Soil column tests under saturated continuous conditions (adapted from Okubo and Matsumoto, 1983).

Chemical clogging

Chemical interactions between infiltrated water and soil leading to clogging include precipitation and dissolution. To illustrate, the application of sodic water to SAT with sodium concentration as low as 5 to 10 % by mass or the application of surfactants caused pore obstruction with dispersed clay due to an increase in Na adsorption and cation exchange capacity (Yaron *et al.*, 2007; Rengasamy *et al.*, 1996; Shainberg and Levy, 1992). Calcium carbonate precipitation and deposition is another clogging factor and is driven by the proportion of divalent cations and hence the direction of the pH. Indeed, a high concentration of divalent cations increases pH and therefore sorption and precipitation (Lindqvist and Bengtsson, 1995). VanGulck and Rowe, (2004), correlated calcium precipitation to acetate fermentation. While Kirk *et al.* (2007) studied kinetics of calcium carbonate precipitation in soil and determined precipitation rate model based on ammonia emission from soil, affecting the soil pH, inhibitors such as organic carbon, phosphate and sulphate, as well as nucleation sites and diffusion. However, other parameters can interfere with this model such as nitrification, carbon dioxide pressure, fungi hyphens and plant roots.

Obstruction of pores also results from inorganic gel formation after calcium dissolution and its reaction with the silicon contained in wastewater (Blazejewski and Murat-Blazejewska, 1997). Due to high occurrence of calcium this is typically more prevalent in limestone areas. In addition, the dissolution of calcium carbonate and other cementing agents leads to soil compaction (Fox, 1999). Blockage of pores by colloids accumulation is another clogging factor to be taken into account. The colloids' surface hydrophobicity may affect colloids retention and adsorption in soil with low carbon concentration. The hydrophobic effect is also enhanced by the dissociation of surface carboxylic groups that increases hydrophobic colloid sorption (Lindqvist and Bengtsson, 1995). The concentration-dependent transport of DOC has been reported by Dunnivan *et al.*, (1992), with slower transport of the hydrophilic fraction than the hydrophobic fraction of DOC.

Table 2-17 Chemical causes and factors reducing soil permeability.

Factor reducing permeability of soil.	Cause
Clay particle size distribution	Increase of sodium adsorption ratio (determined by the ratio of mono to divalent cations)
Sorption, and calcium carbonate precipitation and deposition	Increase of pH and divalent cations, acetate fermentation, ammonia emission and volatilization leading to an increase in pH, high density of nucleation sites
Blockage of pores by inorganic gel formation	Calcium dissolution in limestone and reaction with silicon in wastewater
Compaction of soil	Dissolution of media, like calcium carbonate and other cementing agents
Blockage by colloids retention, sorption, retardation (in soil with low carbon concentration)	Colloids surface hydrophobicity, charges interactions and functional group (ie. carboxylic) dissociation. Slower transport of the hydrophilic fraction than the hydrophobic fraction of DOC.
Desaturation due to gas bubble formation	Hydrocarbon and high concentration in organic compounds leading to supersaturation of methane

Biological clogging

Biological clogging is predominant in systems with prolonged periods of water inundation (Rice, 1974). Prokaryotes and fungi are the predominant micro-organisms in saturated systems but facultative anaerobic bacteria have been found to cause extensive clogging as well (Seki *et al.*, 1996; Shaw *et al.*, 1985). In general there are various mechanisms of biological clogging and depending on the specific physicochemical properties of the soil environment different types become more significant.

The hydraulic conductivity of soil is directly affected by microbial activity and especially secretion of extracellular polysaccharides (EPS), soluble microbial products, proteins and polyuronides (Table 2-18) (Avnimelech and Nevo, 1964; Marshall, 1985; Mitchell and Nevo, 1964; Ragusa *et al.*, 1994; Vandevivere and Baveye, 1992; Allison, 1947). These compounds are slowly biologically degraded, and cause pore clogging because of their ability to fill water-conducting spaces or to enhance cell aggregation (Raiders *et al.*, 1986; Rennie *et al.*, 1954; Shaw *et al.*, 1985; Stewart and Kim, 2004; Vandevivere and Baveye, 1992). This phenomenon is partly controlled by energy source (nutrients), temperature, oxidation-reduction potential, dissolved oxygen, and C:N ratio. The latter influences the ratio of stable to unstable polysaccharides, with a high C:N ratio indicative of resistant polysaccharides (Avnimelech and Nevo, 1964; Frankenberger *et al.*, 1979; Hilger *et al.*, 2000; Vandevivere and Baveye, 1992). For instance, polysaccharides are predominantly produced under anaerobic conditions where their degradation is inhibited (Nevo and Mitchell, 1967). At temperatures below 20 °C EPS degradation seems to be inhibited whereas production continues slowly, and the opposite feature is found above 37 °C (Nevo and Mitchell, 1967). However, a higher temperature, which leads to higher growth rate and activity, can also cause clogging of pores by biomass accumulation but can also enhance degradation of organic matter that was clogging pores (Okubo and Matsumoto, 1983; Platzer and Mauch, 1997).

Furthermore, accumulation of ferrous sulphide colloids acts as a plugging agent in combination with biomass or biological end-products, such as precipitation of metals via sulphate reducing bacteria and other chemotrophic organisms (Baveye *et al.*, 1998; McGauhey and Winneberger, 1964). However, Mitchell and Nevo, (1964), suggested that these colloids do not clog pores directly but are an indicator of reducing conditions that result in polysaccharide accumulation in soil, furthermore they diminish the organic availability for EPS production. Biological reduction of Fe(III) to Fe(II) leads to the formation of iron sludge by precipitation and amalgamation with biological extracellular products (Motomura, 1969).

Another bio-product causing a reduction of permeability is biogas. Entrapment of azoth and methane, produced by micro-organisms, can block pores especially at the pore necks, by occupying up to 30 % of the volume (Baveye *et al.*, 1998; Poulovassilis, 1972; Reynolds *et al.*, 1992; Ronen *et al.*, 1989; Seki *et al.*, 1996; Seki *et al.*, 1998).

Growth, coalescence and flocculation of micro-organisms affect the physical properties of the soil media, including pore volume, size, network and roughness by forming multilayer, coating biofilm on grains, micro-colonies at lower C:N ratios, or simply by accumulation of cells (Baveye and Valocchi, 1989; Cunningham *et al.*, 1991; DeLeo and Baveye, 1997; Molz *et al.*, 1986; Vandevivere and Baveye, 1992). The seed for microbial growth may come from indigenous or injected biomass and growth is aggravated with applied nutrients (Bouwer, 1994; Johnson *et al.*, 1999). Harvey *et al.*, (1984) showed that more than 95 % of the biomass was attached directly to particle surfaces resulting in a filtration cake formation. However, recent studies tend to support the micro-colony model rather than the biofilm hypothesis (Miyazaki, 2006). Moreover, strategic location of biomass growth within pores does not need a great amount of cells to greatly reduce the permeability (Baveye *et al.*, 1998; Characklis *et al.*, 1987).

Immobilization or retardation of micro-organisms is affected by their colloidal properties (i.e. hydrophobicity and charge) and physiological properties (i.e. kinetics, metabolism, shape and size). For instance, flagellated bacteria are adsorbed at a higher rate than non-flagellated bacteria and in general any colloids with steric bulk or surface properties are adsorbed to a greater degree (Stanley, 1983). However, within the group of flagellated micro-organisms, predators of bacteria, such as *Heteromita globosa*, can be found, which are known to maintain soil permeability by grazing on biofilm (Mattison *et al.*, 2002).

Finally, the prevalence of different modes of biological clogging depends on the properties of the media such as the specific surface area available for biofilm growth. Low or reduced permeability media, such as fine grains, is more susceptible to clogging than higher permeability media (Vandevivere and Baveye, 1992). On the other hand, at a high porosity, some fine but porous media can maintain permeability, since clogging occurs preferentially in the largest pores, while biofilms tends to develop within coarse particles rather than on their surface where the specific surface area is less. The chemical characteristics of the rock and groundwater may also promote different microbial communities (Cunningham *et al.*, 1991; Kalish *et al.*, 1964; Raiders *et al.*, 1986; Vandevivere and Baveye, 1992) which affects the magnitude and type of biological clogging. For instance, biodegradation of the organic fraction of the soil leads to the dispersion of soil particles (Martin, 1945).

Table 2-18 Biological causes and factors reducing soil permeability.

Factor reducing permeability of soil.	Cause
Pore clogging because of reduction of water-conducting spaces	Microbial activity, and especially secretion of EPS, SMP, proteins and polyuronide
Pore clogging because cell aggregation is enhanced by EPS (polysaccharide) production	High energy source, low oxidation-reduction potential , low dissolved oxygen, and carbon to nitrogen ratio, low temperature
Pore clogging by accumulation of iron sludge/ colloids	Biological reduction of Fe(III) into Fe(II) which then may precipitate or get combined with biological extracellular products or/and biomass , such as sulphate reducing bacteria (SRB), producing ferrous sulphide colloids
Deterioration of pores volume, size, connexion and roughness of the media	Growth, coalescence and flocculation of micro-organisms to form a multilayer, coating biofilm Micro-colonies at lower carbon to nitrogen ratio
Pore clogging by accumulation of biomass	Immobilization or retardation of micro-organisms is affected by their colloidal and properties. Flagellated bacteria are adsorbed at a higher rate than non-flagellated bacteria and in general any colloids are also strongly adsorbed.
Increase of pressure and desaturation of soil	Entrapment of biogas N_2 and CH_4
Soil particles dispersion	Biodegradation of soil organics

2.4 Wastewater treatment for SAT reuse

2.4.1 Main options of wastewater treatments

As a global approach, the selection of pre-treatment for SAT reuse can be based on the following criteria (Asano and Wassermann, 1980; NRC, 1994; NRC, 1998). The first is the suitability of technology for the required effluent quality, bearing in mind the pollutants present and with regards to the existing treatment plant, nearby SAT performance, reliability and soil characteristics. For instance, tight soils that require vegetative cover cannot be flooded for long periods and therefore necessitate further pre-treatment for denitrification

(Bouwer, 1996). Also, rainfall must be taken into account since it may elute adsorbed endotoxines and viruses (Gerba and Lance, 1978; Goyal *et al.*, 1980; Wellings *et al.*, 1975). Short-term or long-term (depending on soil depth and residence time) SAT will not require the same pre-treatment to achieve potable water quality. For instance, denitrification would be required using short-term SAT for DOC reduction though not with long-term SAT (12 months) (EC, 2006). Secondly, performance and reliability of the whole process and of each barrier for the pollutants present in the influent must be determined and managed by quality assurance procedures. This will allow definition of the environmental impact of SAT reuse systems and the risks to soil and groundwater. The main concern is trace organic compounds, even at below toxic thresholds, since other pollutants are usually well removed by pre-treatment, SAT and post-disinfection (Bouwer, 1996). These recalcitrant chemicals accumulate in the soil until saturation (Bouwer, 1991). The indigenous biomass is also to be preserved since it plays an important role in SAT process such as NOM removal (Langmark *et al.*, 2004). Finally, capital and operating costs are evaluated to determine the project viability and the benefice analysis and depend highly on local costs (land, maintenance, energy) and on the market and the technology development trends.

Assuming an appropriate and efficient SAT site and management, pre-treatment selection needs to focus on the compounds that SAT has limited removal potential for, as well as those shown to cause SAT clogging (Table 2-19). Different technologies have been evaluated as pre-treatments including sand passage, membrane filtration, biological growth and chemical treatment. The best options consist of subsurface flow constructed wetland (SSFCW), conventional activated sludge (CAS) and membrane bio-reactor (MBR). However, multi-barrier systems must be considered for the best performance. For example, a reed bed could be added after AS for further salts, P and pharmaceuticals removal. In the same way, reverse osmosis (RO) will have added value after MBR and SSFCW but requires intermediate treatment after AS. Advanced oxidation processes (AOPs) can also be considered as a tertiary treatment with high removals of metals and pharmaceticals, while coagulation/ flocculation will ensure P and DOC removal including the non-biodegradable fractions.

Table 2-19 Suitability of pre-treatments for groundwater quality and SAT clogging prevention (- 0-25 % removal, + 25-50 %, ++ > 50 %; Cbz Carbamazepine) adapted from Metcalf and Eddy, (2003).

Treatments	Removal of pollutants						Clogging prevention					Cost Capex /Opex*
	DOC	N (NH_4/NO_3)	P	Al, Fe, Mn	Cbz	Tot. *Colif.*	TSS	Salts (TDS/ $CaCO_3$)	DO	Nutrients	Fe	
Primary treatment	+	+/-	-	+	+	-	+	-/-	+	-	+	
Constructed wetland (1)												
- FSF	-/+	+/+	+	++	+	++	++	++	-/+	+	-/+	200/6
- SSF	+	++/-	+	++	+	++	++	++	+	++	++	400/6
Suspended growth												
- CAS (2)	++	++/-	+	-/++	-	++	++	-/+	++	++	++	700/35
- BNR (3)	++	++	++	-/++	-	++	++	-/+	+	++	++	
Attached growth (4)												
- Trickling Filter	+	-/-	-	+/++	-	+	++	-/-	-	+	+	800/100
- RBC	-	++/-	-	+/++	-	-	++	-/-	-	+	++	700/20
Chemical processes												
- coag. Flocc. (5)	++	+/-	++	+/++	+	++	++	-/+	-/+	+	++	
Filtration												
- SF (6)	+	++/-	+	-	-	++	++	-/-	-/+	+	-	150/15
- AC (7)	++	+/-	++	+	+	++	++	-/-	-/+	+	++	200/200
Membrane process												
- MBR (8)	++	++/-	+	-/++	-	++	++	-/+	++	++	++	1500/177
- NF/RO (9)	++	+	++	-	++	++	++	++/-	++	++	-	1200**/114
Advanced treatment												
- AOPs (10)	++	++/-	-	++	++	++	+	+/-	++	+	++	/1000

FSF free surface flow; SSF subsurface flow; CAS conventional activated sludge; BNR biological nutrient removal; RBC biological rotating contractor; SF sand filter; AC activated carbon; MBR membrane bio-reactor; NF nano-filtration; RO reverse osmosis; AOP advanced oxidation process.

1. Park *et al.*, (2008); Lazareva and Pichler, (2009); Matamoros *et al.*, (2007); Paing and Voisin, (2005); Perkins and Hunter, (2000)
2. Radjenović *et al.,* (2007)
3. Rosal *et al.,* (2009)
4. Kasprzyk-Hordern *et al.*, (2009); Kim *et al.*, (2007)
5. Reungoat *et al.*, (2009)
6. Nakada *et al.*, (2007) ; Hamoda *et al.*, (2004)
7. Reungoat *et al.*, (2009)
8. Judd, (2006)
9. Radjenović *et al.* (2007), (2008); Johnson *et al.*, (1999); Sadr Ghayeni *et al.*, (1996)
10. Klavarioti *et al.*, (2009); Poyatos *et al.*, (2009); Caretti and Lubello, (2003)

* Capex based on $1000 per 100 000 gal.d^{-1} flow and Opex based on $1000 per year for this flow adapted from US-EPS (2008)

** Capex based on $1000 per 500 000 gal.d^{-1} flow, Griffin and Gooch, (2009)

2.4.2 SAT and pre-treatment system capabilities

Some studies have shown no difference or even better results in terms of SAT effluent quality and infiltration rate using primary effluent rather than with secondary effluent (Carlson *et al.*, 1982; Ho *et al.*, 1992; Lance *et al.*, 1980; Leach *et al.*, 1980; Rice and Bouwer, 1984). Indeed, higher TOC content leads to a better removal of recalcitrant TOC in the SAT by secondary utilisation and co-metabolism (McCarty *et al.*, 1984) and enhances denitrification (Lance *et al.*, 1980). In addition, if clogging zone development is retarded or absent, for example due to the application of highly pre-treated effluent, removal of pathogens and other constituents of concern is also reduced because this zone is biologically active when present (Baveye *et al.*, 1998). Conversely, if the soil is excessive cloggedthis may cause hydraulic dysfunction, soil anaerobiosis and reduce purification (Van Cuyk *et al.*, 2001). This may lead to conflicts between optimum pre-treatment for SAT and the pre-treatment required by the local authorities, who have to manage as well public acceptance (Bouwer, 1996). Nevertheless, higher levels of pre-treatment may be recommended for virus removal and clogging prevention, since TSS and DOC can lead to physical and biological clogging (Bouwer, 1996). Also, higher pre-treatment levels can insure the controllability and reliability of effluent quality, which is often more difficult with SAT only.

Jimenez and Chávez, (2004) evaluated the feasibility of reusing infiltrated raw wastewater from the Tula Valley, Mexico, for drinking water (Table 2-20). The results showed extensive removal of pollutants and bacteria, however the authors addressed their concern about unknown pollutants and safe use, and suggested the use of further pre-treatment. In India, Nema *et al.*, (2001), also reported satisfactory results using primary treated wastewater followed by SAT for irrigation reuse with 90 % removal of COD, BOD and TSS, 50 % removal of ammonia and a microbial reduction of 4 to 5 log. This study emphasised the importance of the superficial mat for pollutant removal.

Idelovitch *et al.*, (2003), and Kanarek and Michail, (1996), studied an Israeli site in the Dan Region, where SAT is preceded by an activated sludge process modified for nitrogen and phosphorous removal. The performances ranged between 99 and 100 % for TSS, COD, BOD, phosphorus and ammonia, 57 % for total nitrogen, 75 % for DOC and 3.2 log reduction of bacteria. They noticed an increase in manganese, sodium and boron, but a general high degree of reliability and consistency.

Tertiary treatment of wastewater has been more recently considered, since pathogens (enteroviruses and protozoan cysts) and toxic organic compounds from secondary effluent have been recovered in recharged groundwater (Lee and Jones-Lee, 1996). In USA, most of indirect potable reuse projects are using conventional secondary treatment followed by tertiary treatment prior to surface spreading and SAT or tertiary filtration for subsurface injection (Bouwer, 1996; Drewes and Shore, 2001; Drewes *et al.*, 2002). Fox *et al.*, (2006), studied denitrified micro-filtered treatment followed by a saturated anoxic SAT. Despite the high quality effluent of the advanced filtration processes, the effluent ions balance may lead to perturbation of geochemistry of soil (Johnson *et al.*, 1999). Fox *et al.*, (2006), demonstrated that denitrified effluent prior to short-term SAT leads to lower DOC content. However, the higher C:N ratio of primary treated effluent leads also to higher denitrification during SAT.

The removal of some compounds by SAT does not depend on pre-treatment if the SAT capacity is adequate. For instance, it appears that virus adsorption into soil depends only on virus type and soil type and not on virus concentration in the influent water (Blanc and Nasser, 1996).

In developing countries, the application of low level of pre-treatment for SAT has showed economic viability and better effluent quality than available wastewater treatment plants. In addition, the biologically-active zone at the surface of the SAT basin can act as an additional barrier to pollutants and can easily be removed during the drying period to maintain surface permeability.

The examples of SAT systems reported in developed countries represent consistent and reliable systems. However, over the long-term, site specific geochemical and hydrological changes have been noticed driving research projects towards solutions of lesser impact.

Table 2-20 Examples of different levels of SAT pre- treatment and performances (ND not detected).

Level of wastewater treatment prior SAT	Removal [%]	Effluent quality [mg.L^{-1}] [CFU.100mL^{-1} for bacteria]	Reference
Raw	TSS 99	TSS 0.1-2	Jimenez and Chávez, 2004, Tula Valley, Mexico
	TOC 84	TOC 5.2-30	
	BOD 98	BOD 2.4-5	
	NH_4-N 97	NH_4-N ND-4.5	
	TN 97	TN ND-6	
	PO_4-P 95	PO_4-P ND-0.2	
	Faecal *Coliforms* 99.9	Faecal *Coliforms* 1-4	
Primary	BOD 90	BOD 9.3	Nema *et al.*, 2001, Ahmedabad, India
	COD 90	COD 24	
	TSS 90	TSS 18	
	NH_4-N 50	NH_4-N 21	
	bacteria 99.99	PO_4-P 0.2	
Secondary CAS modified for N and P removal	TSS 100	TSS 0	Idelovitch *et al.*, 2003; Icekson-Tal *et al.*, 2003; Kanarek and Michail, 1996; Israel Dan Region
	COD 99	COD 8-11	
	BOD 99	BOD <0.5	
	NH_4 99	NH_4-N 0.03-0.11	
	NO3 nitrification	NO3-N 9.34	
	TN 57	NO_3-N 1.3-5.33	
	DOC 75	PO_4-P 0.01-0.02	
	PO_4 98	PO4-P 0.05	
	total *Coliforms* 100	total *Coliforms* ND	
Tertiary	DOC 77 (SAT removal only)	DOC 1.24	Fox *et al.*, 2001, Mesa USA
Denitrification + tertiary filtration	TN 20-50 (SAT removal only)	TN < 10	
BNR+UF/RO	NH_4 100	NH_4-N ND	Viswanathan *et al.,* 1999; Al-Otaibi *et al.*, 2001, Sulaibiya area, Kuwait
	NO_3 21-60	NO_3-N 2-3.7	
	PO_4 80-99	PO_4-P 0.1-3.3	
	TSS <0 in SAT	TSS 5-2800	
	COD 70-90	COD 2-7	
	total *Coliforms* 99-100	total *Coliforms* ND-80	

2.5 Conclusions

SAT systems provide reliable, effective and inexpensive process for polishing wastewater compared to more energy extensive processes such as RO. The long retention time of contaminants allows biological, chemical and physical removal mechanisms to contribute to providing a high quality treatment system. Unfortunately, clogging factors interact in the same way. Clogging can be due to pore obstruction by dispersed matter, biological and chemical coating, and soil compaction. Nevertheless, a well selected and operated pre-treatment and SAT will allow the control of clogging factors and removal efficiencies. Indeed, an appropriately designed SAT has a high potential as an advanced wastewater treatment. The pre-treatment required is site specific and its selection is a part of a holistic strategy for health, environmental and financial risk management.

2.6 References

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3 COMPARISON OF DIFFERENT WASTEWATER TECHNOLOGIES AND TREATMENT TRAINS AS PRE-TREATMENT FOR SOIL AQUIFER TREATMENT AND IMPACT OF SLUDGE RETENTION TIME

3.1 Introduction

In Europe, studies on upgrading wastewater treatment plants to facilitate reuse have increased due to awareness of sustainability issues and stricter regulations. Meanwhile, in developing countries low cost solutions are needed to increase water supplies and prevent water born diseases (Bixio *et al.*, 2005). Artificial aquifer recharge is being increasingly considered in Europe and many other regions because it provides additional water supplies and reduces saline intrusion, damage to buildings and aquifer oxygenation. By storing reclaimed water, declining groundwater level and resulting land subsidence can be stopped or even reversed (Asano and Cotruvo, 2004; Dillon, 2002; Fox, 1999). However, even if the extensive capacity of SAT to protect groundwater from pollution has been reported, issues related to health risks and public acceptance are still driving institutions to strengthen reuse standards (NRC, 1994). Preventive actions and high standards for reuse are essential to limit the health risks and increase public confidence in the technology and prevent aquifer pollution, as remediation is difficult. Although strict guidelines or drinking water standards are now applied to groundwater recharge, risks persist as some wastewater contaminants with major impacts that are not covered by existing standards (Table 3-1) (Bixio *et al.*, 2005). In addition, deep clogging phenomena affect infiltration rates over the long term and so require increases in footprint of aquifer recharge systems (Fox, 1999). Reuse of wastewater for irrigation also implies risks of groundwater pollution, public acceptance and soil deterioration. However, reuse standards for irrigation are well established and adapted to different levels of human exposure (Metcalf and Eddy, 2002).

The sludge retention time (SRT) is a key operating parameter of activated sludge plants as it controls the food to micro-organisms ratio (F:M) and affects the biomass properties including concentration of MLSS, settleability, foaming, pollutants' removal and biomass kinetics (Metcalf and Eddy, 2002; Huang *et al.*, 2001). For instance, nitrification performance increased rapidly from 1 to 20 d SRT for a given hydraulic retention time (HRT), with COD removal reaching 90 % at 5 d SRT (Urbain *et al.,* 1998; Soriano *et al.,* 2003; Kraume *et al.,* 2005; Zuehlke *et al.,* 2006). It also impacts on full-scale operation through determining the amount of wasted sludge (Metcalf and Eddy, 2002; Takeshi and Yasuhiko, 1991; Han *et al.*, 2005).

In recent years there has been interest in applying membrane filtration as a pre-treatment for wastewater entering SAT and subsequently used for aquifer recharge. This is because membrane bioreactors (MBRs) have many advantages over conventional activated sludge (CAS) for treatment of wastewater. MBRs enable use of higher SRT, thus producing higher mixed liquor suspended solids (MLSS) and less sludge for disposal. MBRs also proved to be more effective for disinfection and removal of organics and nutrients, as well as more robust (Stephenson *et al.*, 2001). However, despite the lower footprint of MBRs, the capital and operational cost remain relatively high.

Also it has been demonstrated that at the same operational conditions, with a high SRT, CAS and MBR provided similar performance (Soriano *et al.*, 1998; Metcalf and Eddy, 2002). This statement remains controversial due to the significant differences in kinetics between the two technologies observed by Smith *et al.* (2003). Indeed, MBRs allow very high or even infinite SRT, but while some authors reported resulting high performance at increased SRT (Muller *et al.*, 1995; Yamamoto *et al.*, 1989; Rosenberger *et al.*, 2002), others have observed a degradation of kinetics and performance (Khor *et al.*, 2006; Pollice *et al.*, 2008). It is commonly recognised that MBRs allow a reduction in sludge production and associated costs. Nonetheless, they can lead to a reduction in oxygen transfer, accumulation of inhibitory compounds and membrane fouling (Muller *et al.*, 1995; Cicek *et al.*, 2001; Yoon *et al.*, 2004).

Multi-barrier systems are often implemented to insure reclaim water meets regulatory standards, especially for purposes such as indirect potable reuse. Technologies that can also reduce deleterious effects on SAT soil have additional benefits as a pre-treatment step. For that reason, vertical flow reed beds (VFRB) are of particular interest as they are soil based treatment systems and hence represent a potential buffering step for SAT without requiring large areas, effluent recirculation or expensive operation (Cooper *et al.*, 1999). Deep-bed filtration is often used to prevent clogging of downstream reuse systems, specifically irrigated soil (Adin, 1987). VFRBs also protect the soil in from accrual and potential release of residual pollutants from the wastewater.

This paper investigates multiple barriers systems involving VFRB, MBR and CAS, at three different SRTs, namely 6, 12 and 20 d SRT. The different options of pre-treatment were evaluated for irrigation and SAT reuse in terms performance, robustness and compliance (Table 3-1). The study links CAS and MBR floc strength and size, and VFRB respirometry and infiltration rate, with efficacy of treatment. The combination of MBR or CAS followed by VFRB, i.e. VFRB(MBR) and VFRB(CAS) was also compared due to the promise of these treatment trains preceding SAT.

Table 3-1 Wastewater reuse standards for SAT and irrigation purposes (Bixio and Wintgens, 2006; EC, 2006; WHO, 2006; WHO, 2003; Kretschmer *et al.,* 2002; Ayers and Westcot, 1985, Pescod, 1992; Asano and Levine, 1998; Rice, 1974)

Parameter	Significance	Polishing Goal for SAT	Irrigation unrestricted	Irrigation moderate restriction	Irrigation severe restriction
Total Suspended solids (TSS) [$mg.L^{-1}$]	TSS can lead to solids deposit in surface or in depth and consequently anaerobic conditions and soil clogging.	<10	<50	50-100	>100
Organic indicator (COD) [$mgO_2.L^{-1}$]	Their biodegradation can lead to oxygen depletion. For irrigation only excessive amounts can cause problems. Low to moderate concentrations are beneficial for humus and pollutants biodegradation.	70-100	<50		<100
Coliforms [$CFU.mL^{-1}$]	*Coliforms* like *E. Coli* are an indicator of microbial and faecal contamination and health risks	<100	<10	<100	<100
Nutrients [$mg.L^{-1}$]	In irrigation they are beneficial nutrient source for humus and plant growth. However, in excessive amounts they can lead to groundwater contamination and overload sorption capacity in SAT soil.	NH_4-N<2 TN 5-40 NO_3 -N<25	NH_4-N<2 TN<5 NO_3 -N <5 TP<2	TN 5-10 NO_3 -N 5-10	TN 10-30 NO_3 -N 10-30 TP<30
Conductivity [$\mu S.cm^{-1}$])	It is an indicator of dissolved inorganics. Extensive content like sodium may cause permeability problems in soil.	<2500	<700	700-3000	>3000
pH	The pH affects metal solubility and alkalinity and structure of soil, and plant growth.	6.5-9.5	6.5-8	6.5-8	6.5-8

3.2 Materials and methods

3.2.1 Pilot plant design and operation

Experiments were carried out at Cranfield University using the primary effluent from the local sewage treatment works and a pilot treatment train involving a

CAS process followed by a VFRB (VFRB(CAS)), a MBR followed by VFRB ((VFRB(MBR)) and a VFRB fed with primary effluent, all running in parallel (Figure 3-1).

The MBR and the CAS were operated under similar operating conditions with an 8 h HRT and successively at 6, 20 and 12 d SRT. These SRT and HRT were chosen because each condition provide different effluent profile, from low to full nitrification and COD removal (Urbain *et al.*, 1998; Soriano *et al.*, 2003; Kraume *et al.*, 2005; Zuehlke *et al.*, 2007). The MBR design included a tubular membrane submerged in a 35 L aerated biomass tank. . The MBR was set up in submerged membrane configuration, with a 50 cm long membrane module composed of 21 tubes of 0.08 µm pore size, and 6 mm lumen diameter, corresponding to 0.2 m^2 membrane surface area. The CAS was composed of a 30 L aerated biomass tank, a 2 L pre-anoxic zone (to control filamentous growth and foaming) and a 7 L clarifier.

The three VFRB were designed and operated based on the model developed by Grant and Griggs, (2001), (Figure 1). The reed beds consisted of 200 L plastic tanks (1 m high with surface area 0.20 m^2) filled with 0.15 m of filter grade sand (0.5-1 mm), 0.15 m of pea gravel (10 mm), 0.05 m of gravel (20 mm) and 0.6 m of gravel (40-50 mm) from the top to the bottom. All beds were planted with *Phragmites australis* and contained an integrated aeration pipe.

The VFRBs were fed with wastewater 8 times per day for 10 minutes. Flows were 125 $mL.min^{-1}$ for the VFRBs following the CAS and the MBR, and 25 $mL.min^{-1}$ for the VFRB fed with primary treated wastewater, given the higher solids and organics load of the source.

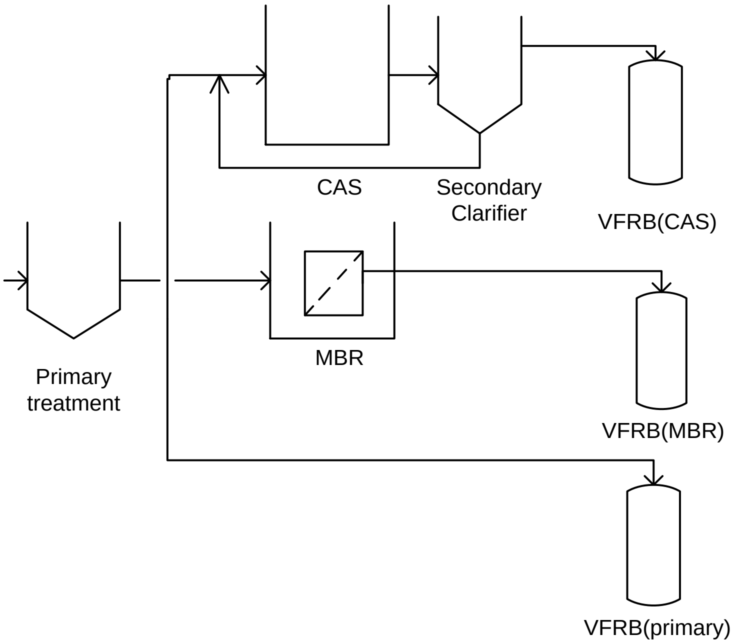

Figure 3-1 Scheme of pilot plant treatment trains.

3.2.2 Water quality analysis

The effluents and influent of the detailed treatment trains were sampled over a period of 18 months after biomass stabilisation (6 months at each SRT) and analysed for total suspended solids (TSS) (after filtration and drying at 105 °C), chemical oxygen demand (COD), ammonia (NH_4), nitrate (NO_3), total phosphorus (TP), total nitrogen (TN) (spectrophotometric test using, Spectroquant cell tests, Merck, VWR International, Poole, UK). As an indicator of the faecal contamination, total *Coliforms* and *E. Coli* count (Compact dry EC, HyServe, Germany), were recorded. Also quantified was sludge volume index (SVI) (standard methods, APHA, 1985). Conductivity and pH were measured using a combined conductivity/pH meter (Jenway 3540, Bibby Scientific Ltd T/As Jenway, Essex, UK). The oxido-reduction potential (ORP) was measured using an ion meter (Jenway 3340, Bibby Scientific Ltd T/As Jenway, Essex, UK). Turbidity was measured using a turbiditymeter (HACH 2100N, Camlab Ltd, Cambridge, UK) after being dispersed for 30 seconds in an ultrasonic bath (Scientific Laboratory Suppliers, Hull, UK).

3.2.3 Activated sludge floc characterisation

The particle size density distribution of flocs from the CAS and MBR processes was calculated using a Malvern Instruments Mastersizer 2000 laser diffractometer (Malvern, UK). Five replicates were used, with a refractive index of 1.33 and a dilution factor of 1:5 for MBR and 1:10 for CAS in order to avoid the flocs agglomeration, based on the dilution factors used by Barbusinski and Koscielniak, (1995). The floc size was calculated from the median value ($d_{0.5}$) after equilibrium at a shear stress of 30 rpm. The steady state floc size at each shear stress between 10 and 100 rpm was recorded to assess floc strength (Biggs and Lant, 2000). Floc strength constants were determined using Equation 3-1 (Leentvaar and Rebhun, 1983).

$$Log d_{\max} = Log C_1 - \gamma Log G \qquad \textbf{Equation 3-1}$$

Where d_{max} is the maximum floc size (m), C_1 the floc strength coefficient, G the average velocity gradient (s^{-1}) and γ the stable floc size constant.

3.2.4 Soil respirometry and organic matter

Aerobic biological activity of reed bed soil was assessed by the soil specific oxygen uptake rate using open respirometry batch tests over 5 d with 4 g of soil of equal moisture content in each cell. Loss on ignition was used to measure of organic matter content of the soils. This test was selected as the VFRB soils had a sandy texture (Rowell, 1994).

3.2.5 Relative robustness of selected treatments

Cumulative percentage value graphs for each effluent quality parameter were used to assess the robustness of the chosen processes. The curves best fitted second order behaviour (Equation 3-2), with regression factors between 0.90 and 1. As the factors A and B described the slope, inflexion point and concavity of the graphs, the ratio of these two factors allowed robustness to be characterised using one number.

$$y = Ax^2 + Bx + c \qquad \textbf{Equation 3-2}$$

3.2.6 Statistics

Results were statistically analysed using ANOVA to determine any significant differences between the treatments trains regarding quality parameters and pollutants' removal. Correlation and determination factors were also used to evaluate if the various parameters tested were correlated. Regression analysis and ANOVA with a confidence level of 95 % were used to confirm or deny the correlation.

3.3 Comparison of membrane bio-reactor and conventional activated sludge process

3.3.1 Water Quality

Organics

COD of MBR effluent at 6 d SRT was 25.9 $mO^2g.L^{-1}$ compared with 36.8 $mg.L^{-1}$ at other SRTs (Table 3-2). Despite this contrast, removal of COD did not significantly vary with SRT as shown by values of 89 %, 92 % and 91% at 6, 12 and 20 d SRTs respectively. COD content of CAS effluent was higher at 12 d SRT than at 6 and 20 d SRT by factors of 1.5, 1.6 and 1.4 respectively. These results were in agreement with an increase in COD in the influent where values were 236, 508 and 329 $mO^2g.L^{-1}$ for 6, 12 and 20 d SRT periods respectively. Also, a lower temperature was observed at 12 d SRT with 5.4 °C compared to 7.2 and 12.2 °C at 6 and 20 d SRT, correlation also found by Grady *et al.*

(1999). This pattern has been observed in activated sludge processes with increased treatment performance as SRT increased up to 8 d, followed by a reduction attributed to the more soluble microbial products (Grady *et al.,* 1999). MBR removed 5 to 10 % more COD than CAS at all SRTs. This suggests retention of particulate or non biodegradable COD by the MBR. These results correspond with those of Hasaand and Kinaci, (2004), who compared CAS and MBR under similar conditions and influent composition as this study and found the MBR produced significantly higher effluent quality. The parameter COD was one of the most affected by SRT variations and overall, it was found the higher the SRT the lower the effluent COD content.

Nutrients

In contrast to the COD results , the CAS effluent quality was similar to or even higher than the MBR effluent quality in terms of NH_4, PO_4 and NO_3 concentration and agreed with other studies under similar same conditions (Urbain 1998; Tchobanoglous, 1991) (Table 3-2). However, TN removal was improved by CAS relative to MBR. This is probably due to the pre-anoxic zone within the CAS system.

Total nitrogen in the MBR effluent at 6 d SRT was 29.8 mg.L^{-1} and thus lower than values of 37.4 mg.L^{-1} at other SRTs. CAS demonstrated a higher TN removal than the MBR, as shown by respective maximum removals of 47 % (12 d SRT) and 33 % (6 d SRT). Phosphate levels were similar following all treatments at all SRTs, with an average of 5 to 10 mgP.L^{-1}. PO_4 removal was highly variable over time and SRT because it depended on influent content.

Ammonium in the CAS effluent (6 and 20 d) and in the VFRB(CAS) train (20 d) were significantly lower with 0.92and 0.355 mgN.L^{-1} compared with values of 2.785 and 1.222 mgN.L^{-1} after 12 d. This was in accord with nitrate levels in the CAS effluent which were higher at 20 d than 6 and 12 d SRT, with respective values of 21.6 mgN.L^{-1} and 12.7 mgN.L^{-1}. The temperature was lower at 12 d SRT than over times, with an average of 5.4 °C and explains the limited nitrification in the CAS trains. Ammonium removal by CAS and MBR ranged between 95 and 96 % at all SRTs. This accord with high ammonium removal reported in literature (Urbain *et al.*, 1998; Soriano *et al.*, 2003; Kraume *et al.,* 2005; Zuehlke *et al.,* 2007).

Solids and bacteria

As expected, membrane filtration was more efficient than CAS for removal of solids and bacteria (Table 3-2). To illustrate, solid contents and turbidity in the MBR effluent were less than 1 mg.L^{-1} and 1 NTU respectively while after CAS equivalent values were 14 mg.L^{-1} and 9 NTU. However, CAS did achieve between 70 and 90 % solids removal and 92 to 97 % turbidity reduction. These results correlate with other studies producing effluent quality of under 2 mg.L^{-1} TSS and 1 NTU for MBRs (Van der Roest *et al.,* 2002; Jefferson *et al.,* 2001; Ng and Hermanowicz, 2005) and 14 to 30 mg.L^{-1} TSS and 3 to 5 NTU for CAS (Massé *et al.*, 2006; Tardieu *et al.*, 1998). Solids content in the CAS effluent after 6 d SRT was significantly higher at 25 mg.L^{-1} compared to an average of 16.7 mg.L^{-1} at the other SRTs. This was related to the CAS floc strength being lower at this SRT (section 3.3.2).

As expected for micro-filtration, only sporadic presence of total *Coliforms* was observed in the MBR effluent at all SRT at levels that can be attributed to the formation of biofilm inside the permeate lines (Côte *et al.,* 1997; Zhang and Farahbakhsh, 2007). Total *Coliforms* content in the CAS effluent was about 3 log higher than after MBR treatment, at 2080-18624 and 0-4 CFU.mL^{-1} respectively. CAS achieved a 0.9-1.9 log removal of total *Coliforms*, similar to other studies (Lucena *et al.*, 2004; Saleem *et al.*, 2000; Zhang and Farahbakhsh, 2007). *E. Coli* was however absent from the MBR effluent compared to an average count of 870 CFU.mL^{-1} in the CAS effluent.

Table 3-2 Effluent quality of MBR, CAS and primary effluent at 6, 12 and 20 d SRT.

Parameters		**MBR**			**CAS**		
SRT	[d]	6	12	20	6	12	20
COD	[mg$_{O2}$.L^{-1}]	26 ± 5	42 ± 9	32 ± 6	49 ± 11	69 ± 10	46 ± 6
TN	[mg$_{N}$.L^{-1}]	30 ± 4	38 ± 4	37 ± 2	33 ± 3	28 ± 5	30 ± 2
NH_4	[mg$_{N}$.L^{-1}]	1.20 ± 0.4	1.00 ± 0.1	0.70 ± 0.5	1.10 ± 0.7	2.80 ± 0.3	0.70 ± 0.6
NO_3	[mg$_{N}$.L^{-1}]	19.8 ± 1.3	34.0 ± 1.8	26.3 ± 3.5	14.3 ± 6.8	11.2 ± 2.1	21.6 ± 3.9
PO_4	[mg$_{P}$.L^{-1}]	4.00 ± 0.6	6.71 ± 0.3	4.81 ± 0.2	4.35 ± 0.4	5.41 ± 1.7	5.32 ± 0.5
TSS	[mg.L^{-1}]	1.0 ± 1.0	1.3 ± 0.6	0.2 ± 0.0	24.5 ± 12.3	14.7 ± 3.5	18.0 ± 10.0
Turbidity	[NTU]	0.7 ± 0.2	0.9 ± 0.1	0.5 ± 0.5	9.0 ± 3.0	9.0 ± 3.4	9.2 ± 2.1
Total *Coliforms*	[CFU.mL^{-1}]	4 ± 4	0 ± 0	3 ± 3	18624 ± 9377	3150 ± 863	2080 ± 1085
E. Coli	[CFU.mL^{-1}]	0 ± 0	0 ± 0	0 ± 0	2068 ± 648	240 ± 33	303 ± 241

Parameters		**Primary effluent**		
SRT	[d]	6	12	20
COD	[mg$_{O2}$.L^{-1}]	236 ± 50	510 ± 490	329 ± 81
TN	[mg$_{N}$.L^{-1}]	44 ± 5	52 ± 14	45 ± 3
NH_4	[mg$_{N}$.L^{-1}]	28.4 ± 2.0	27.8 ± 3.8	27.0 ± 3.5
NO_3	[mg$_{N}$.L^{-1}]	0.1 ± 0.5	0.0 ± 0.0	0.1 ± 0.3
PO_4	[mg$_{P}$.L^{-1}]	4.28 ± 0.7	11.66 ± 2.9	7.81 ± 1.2
TSS	[mg.L^{-1}]	81.0 ± 10.0	141.0 ± 49.8	145.8 ± 40.0
Turbidity	[NTU]	120 ± 40.1	271 ± 119.2	184 ± 27.9
Total *Coliforms*	[CFU.mL^{-1}]	271000 ± 147000	197000 ± 500	169000 ± 25000
E. Coli	[CFU.mL^{-1}]	57300 ± 8900	24000 ± 1000	40200 ± 6500

3.3.2 Floc characterisation

The floc size in CAS was similar at 6 and 12 d SRT with 380 and 400 µm and was slightly lower at 20 d SRT with a floc size of 252 µm (Figure 3-2-A). The MBR floc size was 224, 85 and 39 µm at 12, 20 and 6 d SRT respectively (Figure 3-2-B). The results were in the range of 0.5 to 1000 µm reported in literature. However, CAS flocs size was higher than size of 100 µm typically found by other researchers (Knudson *et al.*, 1982) (Table 3-3). This may be explained by high COD loading in this study. Barbusinski and Koscielniak, (1995), demonstrated a direct proportionality between CAS floc size and COD loading in the range between 0.1 and 0.9 kgCOD $(kgMLSS.d)^{-1}$. Their study showed how an increase in COD loading from 0.17 to 0.5 $kgCOD.(kgMLSS.d)^{-1}$ resulted in an increase in floc size from 121 to 149 µm. In this study, the CAS loading was 0.66 $kgCOD.(kgMLSS.d)^{-1}$ and consequently floc sizes over 149 µm can be expected.

The MBR loading was even greater than the CAS with values up to 0.78 $kgCOD.(kgMLSS.d)^{-1}$. However, flocs size was 85 and 39 µm respectively at 6 and 20 d and thus remained below the range reported by Barbusinski and Koscielniak (1995) (Figure 3-2-A-). Nonetheless, the value of 85 at 20 d SRT compares well with that from Smith *et al.* (2003), who found $d_{0.5}$ of 90 µm for MBR and 500 µm for CAS at 30 d SRT.

Other studies have found floc size below 100 µm with $d_{0.5}$ of 40 µm (Andreadakis 1993). Characterisation revealed an excess of polysaccharides in flocs, with values up to 18 % with resultant biomass starvation. Consequently, the SVI reported by Andreadakis, (1993), with 241 $mL.g^{-1}$ was intermediate between those of CAS and MBR treatments in this study, with respective values of 365 and 159 $mL.g^{-1}$.

In the literature, the effect of SRT is controversial and ranges widely, even at similar SRTs (Table 3-3). In some cases, punctual reduction in floc size was attributed to the accumulation of extracellular products and carbohydrates during CAS treatment (Andreadakis, 1993), while other studies directly observed reduced floc size at lower SRTs for MBR treatment (Holakoo *et al.*, 2007; Lee *et al.*, 2003).

At 20 d SRT, the strength factors, $LogC_1$ and γ, of CAS flocs were respectively 2.5 and 0.09 (Figure 3-3). LogC was slightly lower than the $LogC_1$ value of 2.9 found by Biggs and Lant, (2000). This suggests that CAS flocs had a higher strength than the activated sludge flocs studied by Biggs and Lant, (2000). The calculated value of 0.09 for γ was lower than that of 0.35 found by Biggs and Lant, (2000). The factor γ is an indicator of the mode of floc breakage. Thus the low γ indicates a low tendency for floc breakage, (Biggs and Lant, 2000). This difference can perhaps be explained by the minimum shear rate of 100 rpm used by Biggs and Lant, (2000).

Floc breakage increased at 20 d SRT, with reductions in floc size up to 5.7 and 1.6 fold for MBR and CAS treatment respectively (Figure 3-3). Maximum floc size was obtained in the MBR at 12 d SRT and CAS at 6 and 12 d SRT, with respective values of 224, 380 and 400 µm. This lead to a greater size reduction when shear stress increases to 100 rpm, as bigger flocs tend to have less strength than smaller flocs (Jarvis *et al.*, 2005). This explains the lower floc size of 252 in CAS at 20 d SRT compared to 6 and 12 d with 380 and 400 µm, despite in floc size tends to increase with SRT (Holakoo *et al.,* 2007; Lee *et al.,* 2003).

In this study, variation of SRT did not significantly affect floc size. Similarly, floc size results were not correlated with TSS in CAS effluent. However, floc size was correlated with nitrification. Zhang *et al.*, (1997), found that nitrification increased when floc size diminished since oxygen transfer is promoted in smaller flocs. This relationship is supported by our results for CAS though the opposite was observed for MBR. This was explained by the floc size in the MBR being smaller at 6 and 20 d SRT compared to CAS.

The higher floc size and lower floc strength in the CAS compared to the MBR allow reduced steric hindrance of pollutants adsorbed onto flocs but also leads to better clarification. In the same way, an efficient flocculation provides homogeneous particles of oval shape in the effluent, which have a low filter clogging potential (Adin, 1999; Lawler, 1997). This makes the design of downstream filters, in our case VFRB, SAT or irrigated soil simpler (Adin, 1999; Lawler, 1997). However, the smaller flocs size in the MBR increases pollutants, nutrients and oxygen availability for biodegradation, while it can also increase the specific cake resistance (Adin, 1997; Ji *et al.*, 2008).

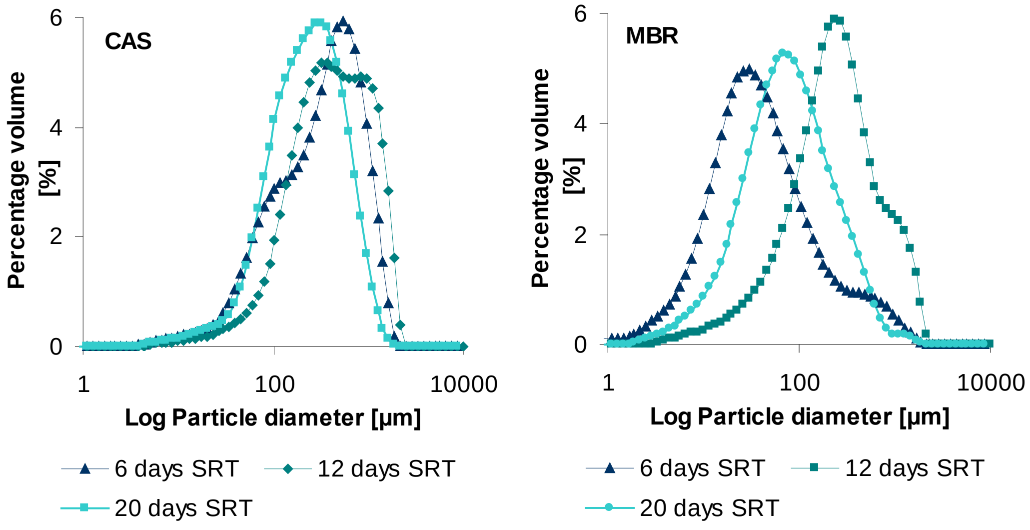

Figure 3-2 Floc size in CAS and MBR at 6, 12 and 20 d SRT.

Table 3-3 Average literature floc size for CAS and MBR at different SRTs

SRT [d]	$d_{0.5}$ [µm]		Reference
	CAS	MBR	
1.1	20		Andriadakis, 1993
3.4	80-90		Zhang *et al.*, 1997
3.6	90-100		
4.2	45		Andriadakis, 1993
6	**380**	**85**	**Pilot plant**
8.8	36		Andriadakis, 1993
8.9	70-80		Zhang *et al.*, 1997
9.7		7-8	
12	**400**	**224**	**Pilot plant**
16.8		20-30	Zhang *et al.*, 1997
17.4	40		Andriadakis, 1993
20	**252**	**39**	**Pilot plant**
20	60-160		Van den Broeck *et al.*, 2009
20		91	Holakoo *et al.*, 2007
20		5.2	Lee *et al.*, 2003
30	428	97.5	Smith *et al.*, 2003
40		135	Holakoo *et al.*, 2007
40		6.0	Lee *et al.*, 2003
60		6.6	
>5 years		30-40	Zhang *et al.*, 1997

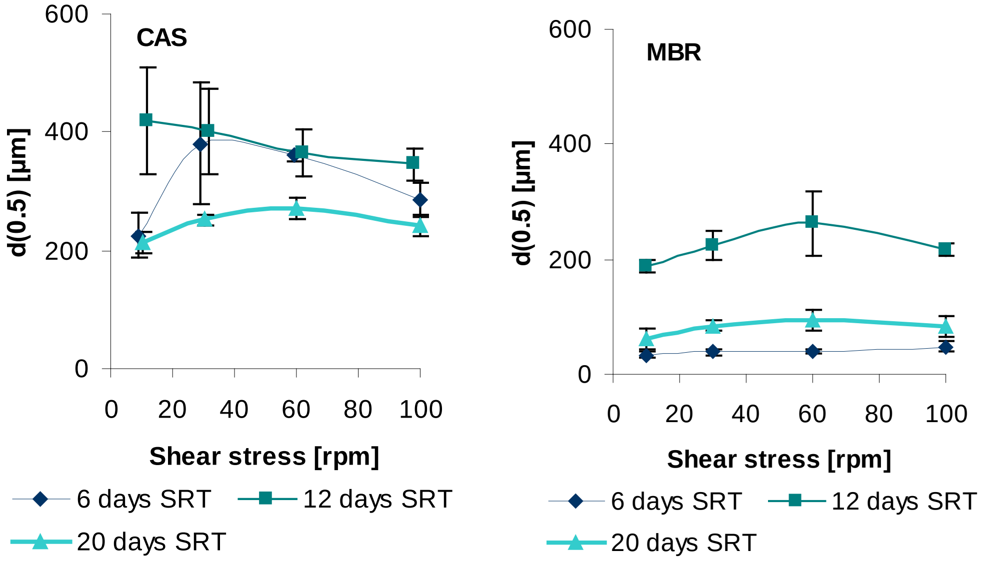

Figure 3-3 Floc strength in CAS and MBR at 6, 12 and 20 d SRT.

3.4 Vertical flow reed beds as secondary and tertiary treatments

In this study, no correlation was found between stage of reed growth or harvest and pollutants' removal. This is in accord with Breen's study, (1997), who observed little effect of VFRB growth stage on removal. This suggests that sand filtration is a significant removal mechanism for VFRB, rather than plant uptake.

3.4.1 Water Quality

Organics

VFRB(primary) achieved good removals of COD, with values between 79 and 88 % and effluent contents between 48 and 59 mgO_2.L^{-1}(Table 3-4). These data are superior to those of with Breen, (1997), with 50-59 % removal and effluent content of 75-79 mgO2.L^{-1}. VFRB(CAS) and VFRB(MBR) provided similar effluent quality in terms of COD for each SRT. As previously seen in CAS effluent, COD content in VFRB(CAS) and VFRB(MBR) effluents was higher at 12 d SRT than at 6 and 20 d SRT by factors 1.6 and 1.4 of respectively corresponding to an increase in COD in the influent and with a lower temperature at 12 d SRT (Grady *et al.,* 1999). The

VFRB(CAS), as a unit, removed more COD than VFRB(MBR), from 40 to 49 % which is close to the range of tertiary VFRB found by Schönerklee *et al.*, (1997), who recorded between 50 and 60 % removal. This compares with COD removal of 0 to 34 % for VFRB(MBR).

Nutrients

Overall, concentrations in N compounds and PO_4 were similar for the three VFRBs, with a few exceptions, while results in terms of removal efficiencies were more variable (Table 3-4).

TN removal in VFRB(primary) was significantly higher during the period at 20 d SRT than during the other periods with 53 % compared to a mean of 27 % during the other periods. At 20 d SRT, VFRB(CAS) provided a higher degree of nitrification than VFRB(MBR), with 25.6 and 20.9 mgN.L^{-1} NO_3 and 0.355 and 0.778 mgN.L^{-1} NH_4 respectively in the effluents. However, a small increase in ammonium was observed in VFRB(CAS) at 6 d SRT and in VFRB(MBR) at 20 d SRT, which can be attributed to ammonification of organic nitrogen (Brix, 1993).

Nitrification of 51 and 52 % was achieved in VFRB(CAS) at 12 and 20 d SRT and of 42 and 73 % in VFRB(MBR) at 6 and 12 d SRT respectively. This agrees with a range of 43 to 91 % found by Schönerklee *et al.*, (1997). In VFRB(primary), nitrification was 95 %, which is above the 80 % reported by Cooper *et al.*, (1997). Nitrification efficiency was lower in tertiary VFRB, corresponding to the lower ammonium content in CAS and MBR effluents.

It was observed that TP was similar in all treatments and for all SRT with an average of 4.80 mg.L^{-1}. Removal of TP was however highly variable over time and SRT. Removal was from for 3 to 46 % in VFRB(primary) treatment to -6 to 25 % in VFRB(MBR) and -7 to 13 % in VFRB(CAS). Variations of a similar magnitude have been previously reported in the literature. For example, an increase in TP has been attributed to the variation in metal ions and release of adsorbed P from soil (Schönerklee *et al.*, 1997; Willadsen *et al.*, 1990).

Solids and bacteria

VFRB(CAS) and VFRB(MBR) obtained a similarly low level of suspended solids, with a mean residual below 10 mg.L^{-1} (Table 3-4). Meanwhile, TSS in VFRB(primary) effluent ranged between 9 and 23 mg.L^{-1}. This demonstrates the high buffering potential of VFRB for CAS effluent treatment. However, this also highlights that VFRB, as anticipated, was degrading the MBR effluent quality in terms of solids as MBR effluent was free from solids. These results tally with literature on the capacity of VFRB for solids, ammonia and COD removals (Cooper, 1999, 2004; Platzer, 1999; Brix and Arias, 2005, Korkusuz *et al.*, 2004). At 12 d SRT, suspended solids in VFRB(MBR) effluent was significantly lower at 1.4 mg.L^{-1} compared to a mean of 6.7 mg.L^{-1} at the other SRTs. Tertiary VFRB provided TSS removal above 90 %, which is higher than removal found by Cooper *et al.,* (1997), who recorded values between 30 and 60 %. Removal in VFRB(primary) was also higher with values consistently more than 80 %. However, TSS effluent content was from 9 to 23 mg.L^{-1} and thus higher than Cooper *et al.,* (1997) who reported 3 to 11 mg.L^{-1}.

Like TSS, VFRB(CAS) and VFRB(MBR) obtained a similarly low level of total *Coliforms* with mean values of 300 and 198 CFU.mL^{-1} respectively (Table 3-4). For VFRB(primary), removal of *Coliforms* ranged from 94 for 99 %. However, only VFRB(CAS) and VFRB(primary) tally with literature for *Coliforms* removal with 79 and 94 % compared to the literature usually between 79 to 99 %, while VFRB(MBR) had negative removal (Wood and Hensman, 1989; Haberl and Perfler, 1990; Výmazal, 1993). This demonstrates once more the high buffering potential of VFRB for CAS effluent treatment. The slight increase in for VFRB(MBR) presumably relates to *Coliforms* originating from the VFRB after cross contamination.

Table 3-4 Effluent quality of VFRBs at 6, 12 and 20 d SRT.

Parameters		**VFRB (MBR)**			**VFRB (CAS)**			**VFRB (primary)**		
SRT	[d]	6	12	20	6	12	20	6	12	20
COD	[mg$_{O2}$.L^{-1}]	28 ± 6	37 ± 8	21 ± 6	27 ± 2	41 ± 13	23 ± 3	48 ± 4	59 ± 5	59 ± 25
TN	[mg$_{N}$.L^{-1}]	29 ± 5	26 ± 3	25 ± 2	27 ± 4	31 ± 5	28 ± 1	34 ± 6	37 ± 7	22 ± 2
NH$_4$	[mg$_{N}$.L^{-1}]	0.7 ± 0.6	0.3 ± 0.0	0.8 ± 0.0	1.3 ± 1.1	1.4 ± 0.5	0.4 ± 0.1	1.5 ± 1.7	1.4 ± 0.7	1.1 ± 0.5
NO$_3$	[mg$_{N}$.L^{-1}]	23.4 ± 3.9	24.9 ± 4.4	20.9 ± 2.9	21.4 ± 2.3	25.3 ± 3.8	25.6 ± 2.9	20.7 ± 7.2	26.7 ± 8.9	17.1 ± 2.7
PO$_4$	[mg$_{P}$.L^{-1}]	4.2 ± 0.4	5.0 ± 0.7	4.5 ± 0.4	3.9 ± 0.6	5.8 ± 0	4.6 ± 0.3	4.1 ± 0.5	6.3 ± 2.2	5.4 ± 0.7
TSS	[mg.L^{-1}]	6.3 ± 3.9	1.7 ± 1.6	8.6 ± 4.0	4.5 ± 1.3	4.3 ± 3.1	8.6 ± 3.8	16.3 ± 6.4	9.1 ± 5.6	22.6 ± 8.5
Turbidity	[NTU]	1.9 ± 0.7	1.6 ± 0.1	0.9 ± 0.2	1.6 ± 0.4	2.1 ± 0.4	2.7 ± 0.2	7.1 ± 4.2	5.2 ± 2.8	7.0 ± 1.8
T. *Coliforms*	[CFU.mL^{-1}]	276 ± 231	105 ± 9	214 ± 40	238 ± 117	235 ± 30	429 ± 126	17152 ± 1633	2663 ± 279	10725 ± 8213
E. Coli	[CFU.mL^{-1}]	6 ± 0	10 ± 0	45 ± 8	35 ± 21	57 ± 28	63 ± 1	2865 ± 304	869 ± 279	3432 ± 2140

3.4.2 VFRB operation

VFRB(primary) demonstrated significant variations over time for soil organic content and respiration, despite the fact that no significant change in effluent quality was observed (Figure 3-4). Soil organic content increased in VFRB(primary) and VFRB(CAS) during the first year of operation corresponding successively to the 6 d and 20 d SRT periods, while all VFRB soils experienced a diminution in organic content after one year when set up at 12 d SRT (Figure 3-4-A-). In the case of VFRB(primary), an accumulation of humus and sludge on the surface is presumably responsible for the increase in organic content and explains the decrease in aerobic respiration caused by reduced soil oxygenation at the 20 d SRT period (Figure 3-4-B-). During the 20 d SRT period, the temperature average was significantly higher than during the 6 and 12 d SRT periods, at 12.2, 7.8 and 12.2 °C respectively. It is probable that proliferation of anaerobic bacteria and deposition of anaerobic by-products deposit occurred during the hottest period, leading to increased organic content not detected through aerobic respiration tests (Blazejewski and Murat-Blazejewska, 1997; Langergraber *et al.*, 2003). The following period was colder with an average temperature of 5.4 °C and probably led to micro-organisms die-off, restoring aeration and reducing the organic content.

For VFRB(CAS) and VFRB(MBR), the higher temperature at 20 d SRT also increased the bacterial activity, leading to an increase in organic content and aerobic respiration (Figure 3-4), but without any surface sludge mat, the microbial community remained predominantly aerobic.

The amount of respiration in VFRB(MBR) did not correlate with nutrients removal or nitrification, while respiration in VFRB(CAS) was in accord with for nutrients' removal and nitrification. This is explained by the relationship between higher aerobic activity and higher removal. For VFRB(primary), the higher TN removal at 20 d SRT corresponded to the lowest level of respiration and the higher organic content of the influent is in agreement with the anaerobic conditions needed for denitrification.

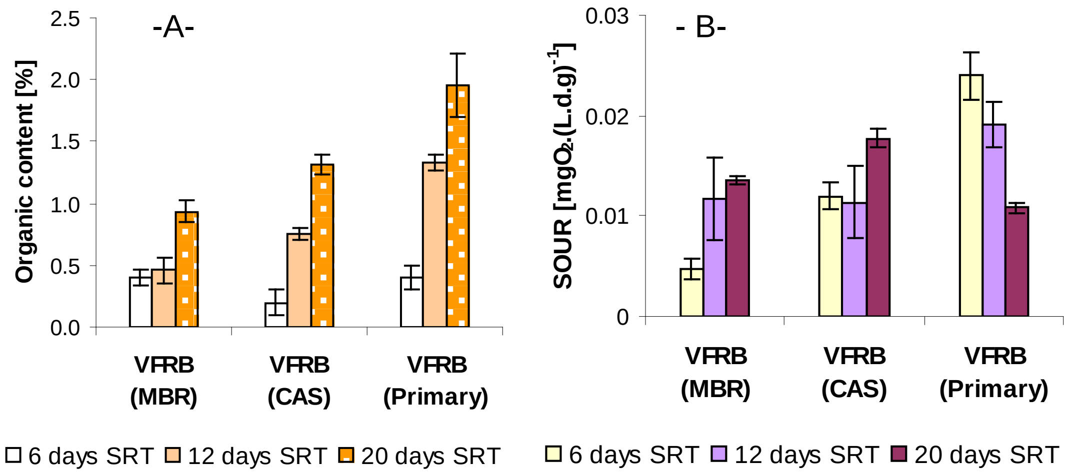

Figure 3-4 -A- Organic content and -B- aerobic specific oxygen uptake rate of VFRB soils.

Over the period of experimentation, no significant soil clogging was observed and the aeration remained stable with a mean effluent DO of 7.29 ± 0.01 mg.L^{-1} in the three VFRBs. Only a slight sludge mat covered the surface of the VFRB fed with primary effluent. It is believed that sludge mats do not influence infiltration rate (Korkusuz, 2004).

The infiltration profile was representative of non clogged soil (Platzer and Mauch, 1997) and was similar for the two tertiary VFRBs with a sharp infiltration rate peak of 250 mL.min^{-1}, 30 min after the feeding started (Figure 3-5). Kayser and Kunst, (2005), observed a similar peak up to 50 mL.min^{-1} after 30 min feeding with lagoon effluent at a hydraulic loading comparable to the tertiary VFRB (204 L.(m^{2}.d)$^{-1}$). The elution peak in VFRB(primary) at a flow of 100 mL.min^{-1} matches that from Kayser and Kunst, (2005), despite their hydraulic loading being more than twice of the VFRB(primary) loading. The flow in the VFRB fed with primary effluent was lower than the tertiary VFRB for design considerations, consequently the water head was lower which and explains the infiltration rate being half of the tertiary VFRBs. The difference in maximum infiltration rates between Kayser and Kunst’s study, (2005), and VFRB in this study can be explained by the differences in media. In Kayser and Kunst’s study VFRB was designed with 20 cm gravel (2-8 mm) on top, 50 cm washed sand

(0-2 mm) and 10 cm gravel (2-8 mm) at the bottom and the influent taken from wastewater lagoons.

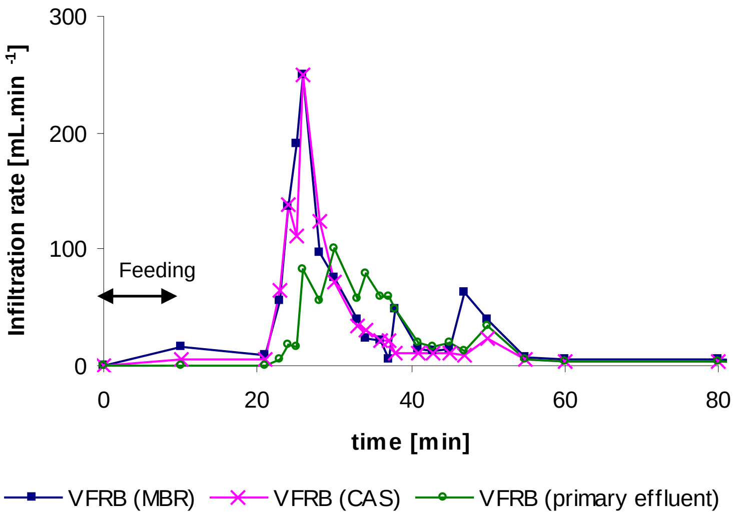

Figure 3-5 Infiltration rate in vertical flow reed beds during wetting periods after 6 months of operation.

3.5 Comparison of treatment trains' robustness

This study demonstrated that the MBR was twice as robust for solids, five times more for *Coliforms*, 10 % more for nitrates and 30 % more for phosphorus than CAS (Figures 3-6 -B- and -C-). However, the robustness of MBR and CAS was similar for COD and nitrogen, and CAS was even 17 % more robust than MBR for ammonia (Figure 3-6 -A-). The higher robustness of the MBR relative to the CAS is in agreement with the literature (Judd, 2006) as CAS is more subject to activated sludge problems such as bulk or filamentous excess. The MLSS was lower in MBR than in CAS and hence not the driver for robustness observed in this study (Metcalf and Eddy, 2002; Stephenson *et al.,* 2000).

Overall, the MBR was also more robust than the three VFRBs for solids, TN and *Coliforms* in effluent. However, VFRB(primary) was the most robust for NH_4 and NO_3 contents, VFRB(CAS) was the most robust for COD and VFRB(MBR) was the most robust for TP.

All VFRBs provided similar robustness for total *Coliforms*, COD, TN and solids in effluent (Figure 3-6 -A- and -C-). VFRB(MBR) was twice more robust than VFRB(CAS) and VFRB(primary) for phosphorus although removals were less. Similarly, VFRB(primary) was five times more robust than VFRB(MBR) and twice as robust than VFRB(CAS) for ammonia, while ammonia content in VFRB(primary) effluent was significantly higher than in the other VFRBs (Figure 3-6). Finally VFRB(CAS) and VFRB(primary) were twice more robust for nitrate content in effluent than VFRB(MBR).

Overall robustness for all SRT ranked in this order: VFRB(primary), MBR, CAS, VFRB(CAS) and VFRB(MBR). Indeed, VFRB(primary) was the most robust for ammonium and nitrates and CAS showed the highest robustness for TN at 12 and 20 d SRT. Nevertheless, MBR was the most robust process at all SRT for *Coliforms* and solids followed closely by VFRB(MBR) and VFRB(CAS) at 12 and 20 d SRT. Despite tertiary VFRB ranked last in terms of robustness, the variations remained at lower concentration than CAS or VFRB(primary).

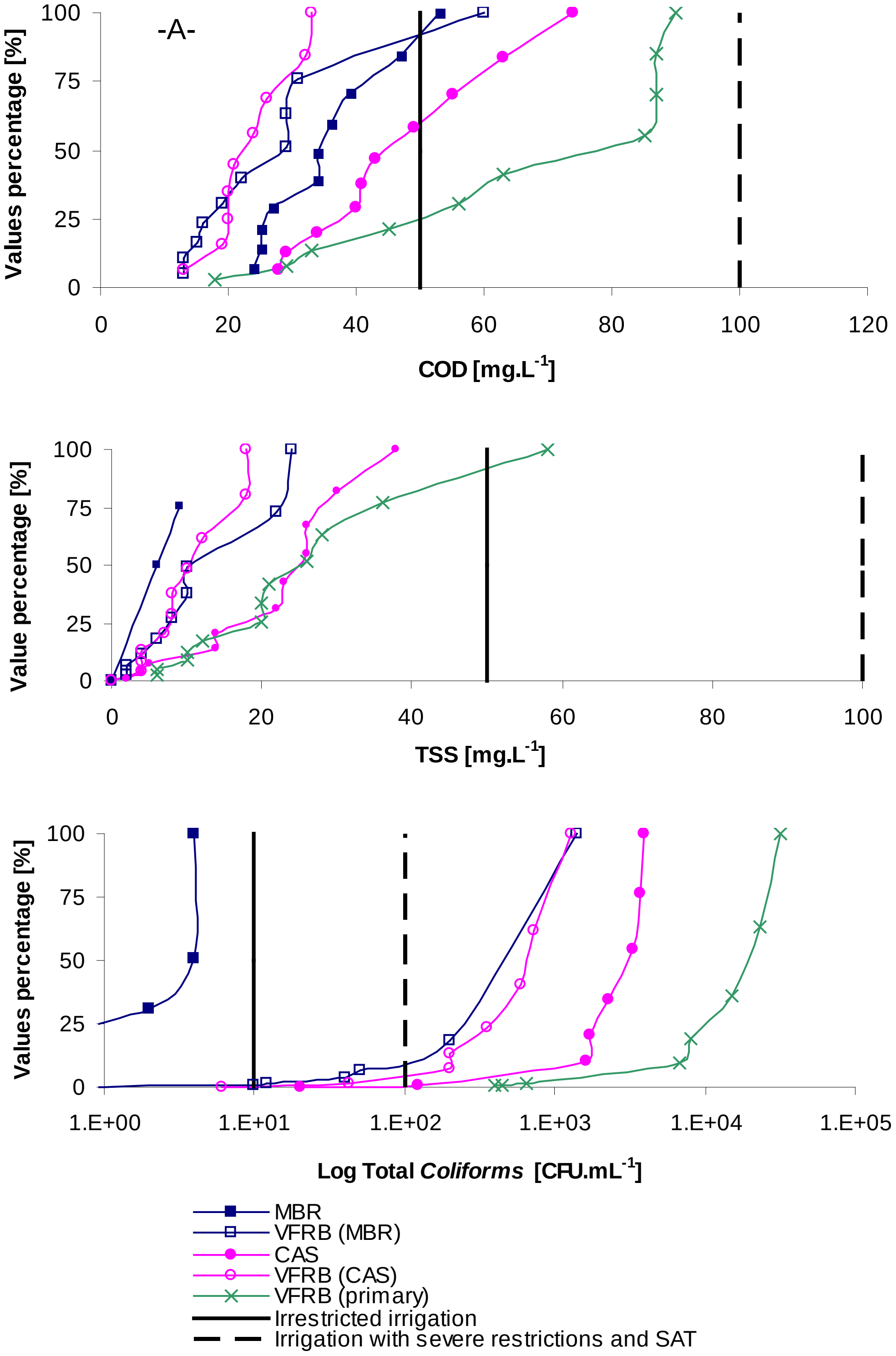

Figure 3-6 Percentage value graphics of -A- COD, -B- TSS and -C- Total *Coliforms* with reuse criteria at 20 d SRT.

3.6 Comparison of treatment trains compliance towards SAT and irrigation reuse standards

The most apparent feature differentiating the treatment trains was the faculty to remove particulate matter. Indeed, bacteria and solids were almost completely removed with the MBR's micro-filtration and well attenuated with VFRB's sand filtration compared to floc adsorption and sedimentation with CAS. This feature impacted the other pollutants as well considering the proportion of particulate organics, nitrogen and phosphorus compounds.

In regard to SAT and irrigation reuse standards, none of the treatment trains offered complete compliance (Table 3-5). Nevertheless, MBR set up at 6 d SRT provided the best option for SAT and restricted irrigation reuse with 95 and 88 % compliance respectively. The second best option was VFRB(MBR) at 12 d SRT where equivalent compliance was 92 and 93 %, albeit with lower robustness than compared the treatment trains ranked fourth and fifth.

At lower SRT, effluents generally had better compliance for nitrate content. For instance, MBR complied at 90 % at 6 d SRT compared to 0 % at 12 and 20 d SRT while still complying for ammonium content at all SRTs. Overall, tertiary VFRB showed higher TN compliance than the other processes up to 100 %. In addition, VFRB(CAS) showed improved compliance for solids and bacteria but reduced compliance for nitrates, while variable effects were observed for pH and conductivity. VFRB(MBR) reduced the compliance of MBR effluent for bacteria and solids but increased it for TN, nitrates, pH and conductivity. VFRB after MBR offered some advantages despite degrading MBR effluent quality in terms of bacteria and solids. Also VFRB(CAS) offered a compliance percentage close to the MBR and VFRB(MBR), at the higher SRTs, with better robustness than VFRB(MBR).

When considering in more detail the best conditions for each treatment, the SAT standard for COD was reached by all the treatment trains (Table 3-5). Previously it has been reported that the pre-treatment do not have any major impact on aerobic biodegradation in SAT (Kopchynski *et al.*, 1996). In addition, for irrigation purposes, moderate concentrations of COD can be beneficial as fertiliser and for soil equilibrium.

MBR effluent was free from solids and bacteria and hence would be the best option for unrestricted irrigation with 100 and 90 % compliance for these two parameters (Table 3-5). This is in accord with Pollice *et al.*, (2004), who demonstrated that the effect of MBR effluent on soil and crops was as good as groundwater. Moreover, bacteria content is very important for irrigation and indirect potable reuse as it is a major health concern (Rose and Clark, 1986). However, even if SAT can easily remove bacteria, they can cause SAT soil clogging, while solids usually lead only to reversible surface clogging (Baveye *et al.*, 1998; Aharoni and Cikurel, 2005).

Tertiary VFRB was beneficial for reuse showing higher compliance than secondary VFRB for all reuse purposes except unrestricted irrigation (Table 3-2). Despite the VFRB degraded MBR effluent quality for solids and bacteria, it showed better overall compliance towards restricted irrigation mainly because of total nitrogen and pH (Table 3-5). VFRB also improved drastically CAS effluent quality, increasing compliance of effluent quality of about 10 %.

Table 3-5 Best compliance percentage towards reuse criteria [%], in reference to Table 3-1 for each parameter (irrigation unrestricted: level 1, with moderate restriction: level 2 and with severe restriction: level 3).

		Treatment trains						
		MBR	**VFRB (MBR)**	**CAS**		**VFRB(CAS)**		**VFRB (primary)**
Parameters	**Reuse Criteria**	**6 d SRT**	**12 d SRT**	**6 d SRT**	**12 d SRT**	**6 d SRT**	**20 d SRT**	**(first 6 months)**
TSS	SAT	100	100	20		90		30
	Level 1	100	100		100		100	100
	Level 2-3	100	100		100		100	100
COD	Level 1	100	100		20	100	100	60
	Level 3 and SAT	100	100	100	90	100	100	100
Total	Level 1	30	70		0	40	0	20
Coliforms	Level 2-3 and SAT	90	75	10	20	40	25	40
TN	Level 1	0	0		0		0	0
	Level 2	0	0		0		14	0
	Level 3	45	90		60		100	36
	SAT	100	100	91		100		73
TP	Level 1	0	0	0	0	0	0	0
	Level 3	100	100	100	100	100	100	100
NO_3-N	Level 1	0	0		0		0	0
	Level 2	10	0		60		0	20
	SAT	90	60	100		90		70
	Level 3	90	70		100		80	100
NH_4-N	Level 1 and SAT	100	100	90	44	80	100	70
pH	Level 1 2 3	80	100		100		100	90
	SAT	80	100	90		90		90
Conductivity	Level 1	40	50		10		0	50
	SAT	100	100	100		100		100
	Level 2	100	100		100		100	100
	Level 3	100	100		100		100	100
					average			
	SAT	95	92	75		86		72
irrigation	level 1	50	58		30		44	43
	level 2	76	75		68		71	69
	level 3	89	93		79		89	82
Robustness	rank per SRT	4	5	2	1	3	4	1

3.7 Conclusion

The study highlighted the benefits of tertiary treating VFRB as a fair and robust alternative to the expensive MBR for restricted irrigation and SAT reuse. Tertiary treatment including VFRB offered as well an additional barrier to pollutants with a high buffering capacity. VFRB after MBR provided good compliance towards reuse standards despite it degraded MBR effluent for solids and bacteria. VFRB(CAS) performances was competitive to MBR and VFRB(MBR) and with a better robustness than VFRB(MBR).

This study also confirmed the role the effect of SRT in CAS on floc properties, organics and nitrification. However, as nitrification increased, compliance towards nitrates was reduced with SRT increasing.

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4 IMPACT OF PRE-TREATMENT AND THEIR OPERATION ON SOIL AQUIFER TREATMENT

4.1 Introduction

Soil aquifer treatment (SAT) systems are used as a wastewater polishing step or complete treatment to avoid a direct discharge of wastewater to water bodies. Indeed, SAT allows reusing water, replenishing groundwater and storing temporarily the restored water in an aquifer (Crites *et al.*, 2006). As wastewater availability is often predictable irrespective of seasonal and climatic variation, it has a great potential as a source of reclaimed water (NRC, 1994). In Europe, over 200 projects of wastewater reclamation were identified by Bixio *et al.*, (2005), though only a small proportion of wastewater reuses is SAT. The SAT process is essentially a low-technology and simple means of providing an advanced and robust wastewater treatment system (Bouwer, 1991). Amy *et al.*, (1993), reusing secondary wastewater effluent, showed that the quality of SAT treated effluent was equal to or even better than that of a conventional tertiary wastewater treatment plant. Indeed, where hydro-geological conditions allow groundwater recharge from surface infiltration facilities, resulting costs are less than 40 % of the costs of equivalent in-plant treatment (NRC, 1994). However, many projects of indirect potable reuse have failed due to the perceived health risk and now high quality of reclaimed wastewater effluent is required also to prevent aquifer pollution (NRC, 1994). Since pathogens and toxic organic compounds from secondary effluent have been observed in recharged groundwater, tertiary treatment of wastewater has been more recently considered to feed SAT and water reuse schemes (Lee and Jones-Lee, 1996). Consequently, multi-barrier systems and advanced treatments such as membrane filtration have been envisaged in more recent projects. However, the effluent ions balance from advanced filtration processes may lead to perturbation of the soil geochemistry despite the high quality effluent (Johnson *et al.*, 1999). As a result, the design of SAT pre-treatment should consider different technologies and their operating conditions in combination with soil characteristics to prevent contamination and irreversible clogging. This paper focuses on the comparison of different treatment trains, from conventional to advanced technologies and as single unit or multi-barriers system, on SAT soil and effluent quality. It also looks at the quality of the effluent obtained after one metre of SAT for potable reuse. This builds on the previous paper which compared the different pre-treatments in terms of compliance towards SAT and irrigation standards and highlighted the benefits of MBR for bacteria and solids removal but also VFRB as a fair and robust alternative to the expensive MBR (Table 4-1).

Table 4-1 Effluent quality and removal efficiencies in SAT pre-treatment.

Parameters	MBR	CAS	VFRB (MBR)	VFRB (CAS)	VFRB (primary)	Primary effluent
COD						
mgO$_2$.L^{-1}	26-42	41-49	21-37	23-41	43-59	236-508
(%)	(89-92)	(79-86)	(88-93)	(88-93)	(79-88)	
NH$_4$-N						
mg.L^{-1}	0.65-1.24	0.75-2.78	0.27-0.78	0.35-1.35	1.12-1.50	27-28
(%)	(96-98)	(90-97)	(97-99)	(95-99)	(95-96)	
TSS						
mg.L^{-1}	0-1.3	14.7-24.5	1.7-8.6	4.3-8.6	9.1-22.6	81-146
(%)	(96-99)	(70-90)	(92-99)	(94-97)	(80-93)	
Total *Coliforms*						
CFU.mL^{-1}	0-4	2-20 .10^3	105-276	235-429	3-20 .10^3	150-300 .10^3
(%)	(100)	(93-99)	(99-100)	(99-100)	(94-99)	

4.2 Materials and methods

4.2.1 Pilot plant

The installation includes a conventional activated sludge process (CAS) followed by vertical flow reed bed, a membrane bioreactor (MBR) followed by vertical flow reed bed (VFRB), a VFRB fed with primary effluent and a SAT soil column following each unit (Figure 4-1). MBR and CAS were running at 8 h HRT and successively 6, 20 and 12 d SRT.

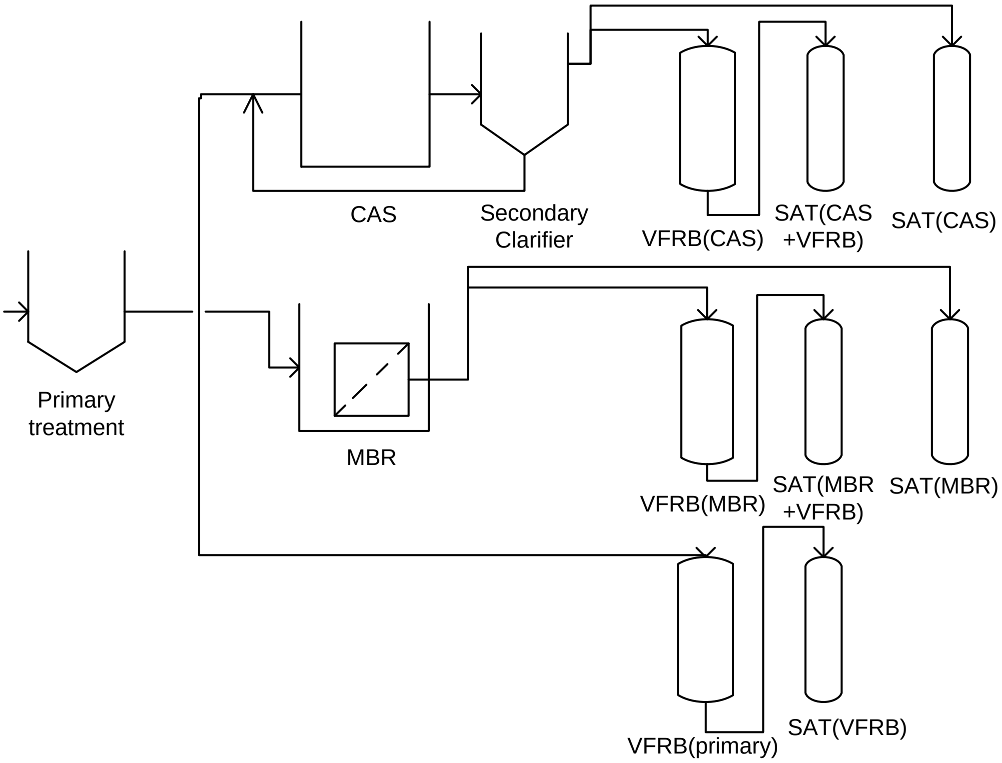

Figure 4-1 Scheme of pilot plant treatment trains.

The SAT columns were operated under unsaturated conditions, a constant head of 5 cm and a rotation of 7 d wetting and 7 d drying periods. Columns were made of clear and rigid PVC pipes and fitting (Figure 4-2). The dimensions are 1.3 m length with an internal diameter of 2.6 cm which allows the approximation of the flow rate to one dimension with negligible wall effect. Columns were filled with 10 cm of gravel and 1 m of sieved (2 mm mesh screen) and repacked aquifer material at 1.5 g.cm^{-3} density. The aquifer material was provided on the courtesy of the Shafdan SAT site, Israel.

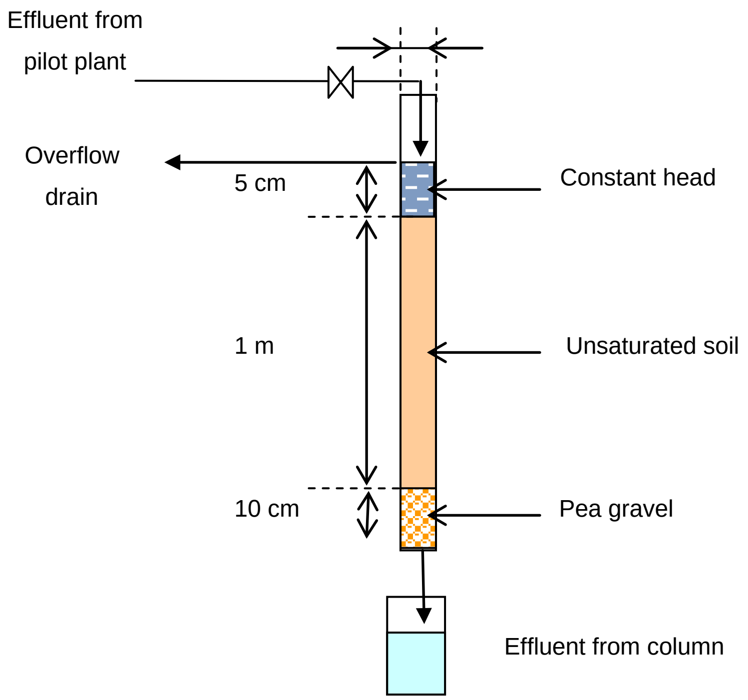

Figure 4-2 Scheme of SAT soil column.

4.2.2 Water quality analysis

The treatment trains effluents and influent were sampled over 18 months (6 months per SRT). The results for VFRB(primary) are not related to SRT but refer to three successive periods of 6 month corresponding in this order to 6, 20 and 12 d SRT in the other treatments that include activated sludge. Effluents were analysed for TSS (filtration and drying at 105 °C), chemical oxygen demand (COD), ammonium (NH_4), nitrates (NO_3), total phosphorus (TP), total nitrogen (TN) (spectrophotometry, Spectroquant cell tests, Merck, VWR International, Poole, UK), total *Coliforms* and *E. Coli* count (Compact dry EC, HyServe, Germany), as an indicator of the faecal contamination, and sludge volume index (SVI) (standard methods, APHA, 1985). The pH and conductivity were measured using a conductivity/pH meter (Jenway 3540, Bibby Scientific Ltd T/As Jenway, Essex, UK).

Analysis of SAT soils recovered after each SRT period were also undertaken using a scanning electron microscope coupled with X-ray diffraction (XL-30

ESEMR Phillips, The Netherlands) to identify any possible changes at the surface of soil grains resulting from either chemical precipitation or biological clogging.

4.2.3 Statistics

Following the sampling campaign, results were statistically analysed using ANOVA to determine any significant differences between the treatments trains regarding listed analytes and pollutants removal. Correlation and determination factors were also used to find out if the various parameters tested in the treatment trains were correlated, subsequently regression analysis and ANOVA were used to confirm or deny the correlation.

4.3 Effect of sludge retention time on pollutant removal in soil aquifer treatment

4.3.1 Organics

COD concentration remained between 18 and 26 mg.L^{-1} for all treatments effluents and at all SRTs except in the case of SAT(CAS) where the COD was higher at 38 and 34 mg.L^{-1} at SRT of 6 and 12 days respectively (Figure 4-3). This can be explained by the CAS effluent having a higher fraction of particulate COD and hence requiring long term retention to transform the recalcitrant fraction (Kopchynski *et al.*, 1996; Fox *et al.*, 2001). COD removal in SAT units accounted for 44 to 60 % in SAT(VFRB), 8 to 48 % in SAT(MBR), 5 to 40 % in SAT(MBR+VFRB), 23 to 55 % in SAT(CAS) and 13 to 38 % in SAT(CAS+VFRB), with the lowest removal usually found at 6 d SRT. These results are lower than those reported by Bouwer *et al.*, (1980), who found COD reduction of almost 100 % by SAT fed with secondary effluent. Concentration of COD in SAT influent was lower in this study and the columns used were three times smaller than in Bouwer *et al.* study (1980).

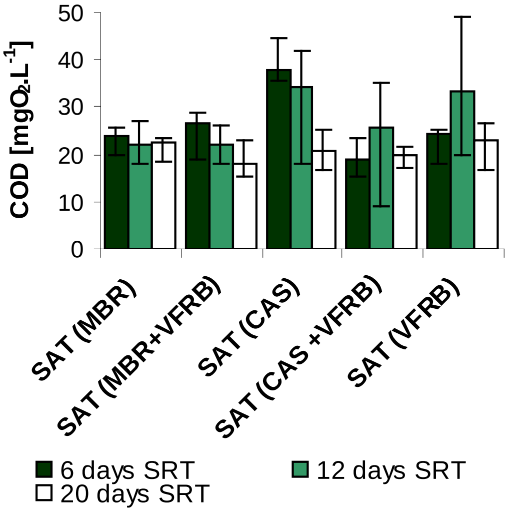

Figure 4-3 COD content in SAT effluents at 6, 12 and 20 d SRT.

4.3.2 Nitrogen compounds

TN removal was higher in SAT(CAS) and SAT(MBR) at 6 d SRT relative to 12 and 20 d, respectively with 13.1 mg.L^{-1} compared to 31.7 mg.L^{-1} for SAT(CAS) at 6 d and higher SRT and 22.4 mg.L^{-1} compared to 34.8 mg.L^{-1} for SAT(MBR) (Figure 4-4). With the exception of SAT(CAS), where 70 % TN removal was achieved, SAT removed less than 50 % TN in accord with the literature when conditions for conventional denitrification are absent (Suzuki *et al.,* 1992). As conventional denitrification was not expected because of the low carbon to nitrogen ratio of 1.5 in CAS effluent at 6 d SRT, the higher TN removal in SAT can be explained by autotrophic denitrification, such occurs during ANAMMOX process and sulphur-limestone process (Fox *et al.*, 2006; WHO, 2004). An alternative explanation is that denitrifying bacteria used the soil carbon (Gable and Fox, 2000).

Ammonium content in SAT(CAS) effluent was higher at 6 d SRT with 0.778 mgN.L^{-1} compared to 0.361 mgN.L^{-1} in average at 12 and 20 d SRT. Conversely, nitrate content was lower at 6 d than 20 and 12 d with respective concentrations of 18.2, 26.1 and 36.0 mgN.L^{-1}, this matching TN removal (Figure 4-5). Ammonium removal ranged from 16 to 95 % for all processes with lower removal at 6 d SRT especially for SAT(MBR) but usually above 60 % for the other SATs. The results were generally lower than Suzuki *et al.,* (1992), who found about 90 % removal of ammonium when applying primary effluent onto SAT.

A small amount of nitrification was observed in SAT units, up to a maximum of 10 % for SAT(VFRB). This corresponded to the removal of remaining ammonia, especially at 6 d SRT when minimal removal was achieved upstream. Only SAT(CAS) and SAT(MBR) did not increase nitrate content at 6 d SRT. Nitrate content in SAT(MBR) and SAT(CAS) was reduced respectively with 19.8 mgN.L^{-1} at 6 d SRT compared to 31.0 mgN.L^{-1} in average at the other SRTs, and 10.8 mgN.L^{-1} compared to for SAT(CAS) at 6 d SRT compared to an average of 31.1 mg$_N$.L^{-1} at 12 and 20 d SRT. Conversely, nitrate content in SAT(MBR+VFRB) was higher at 12 d SRT with 28.3 mgN.L^{-1} compared to 22.7 mgN.L^{-1} at the other SRTs.

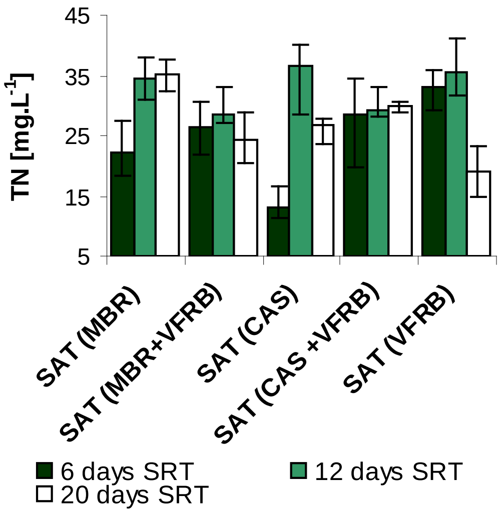

Figure 4-4 TN content in the effluents at 6, 12 and 20 d SRT.

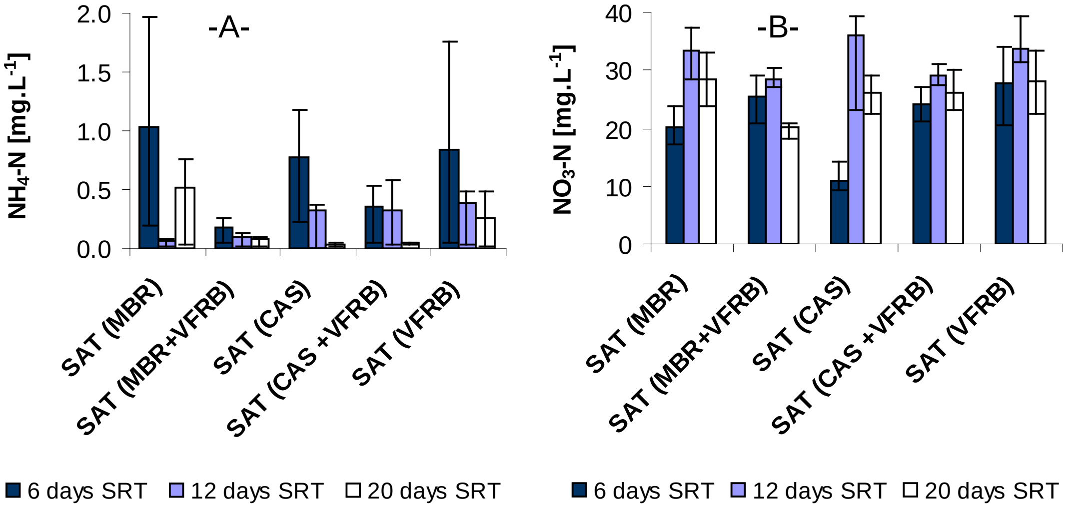

Figure 4-5 -A- Ammonium and -B- nitrates in SAT effluents.

4.3.3 Phosphorus

At 6 d SRT, with the exception of SAT(CAS), TP content ranged between 2.80 and 1.98 mg.L^{-1} and removal ranged between 36 and 49 % for SAT(MBR) and SAT(VFRB) respectively. However, removal below 5 % was observed at 12 and 20 d SRT for all SAT (Figure 4-6). The 6 d SRT results tally with those of Kanarek *et al.*, (1993), and Lin and Banin, (2006), who observed about 50 % phosphorus removal with long retardation factors and breakthrough times. At 6 d SRT, SAT(CAS) effluent only content 0.09 mg.L^{-1} TP. This could be connected to the low infiltration rate of 3.5 cm.d^{-1} allowing the slow processes of phosphorus mineralisation and sorption to occur. For the other SATs, infiltration rates were around 20 times higher. This was also related to the high pH of 8.3 found in SAT(CAS) at 6 d SRT that can enhance or be a result of phosphorus precipitation. The pH was 7.8 in the other SATs and SRTs. In general, SAT increased the pH to an average of 7.8 compared to 6.9 in the pre-treatment effluents. However, subsequent phosphorus leakage was observed after all SAT when changing the SRT from 6 to 20 d SRT. No such trend was measured in influent phosphorus content, thus suggesting the modification or the saturation of the soil sorption capacity.

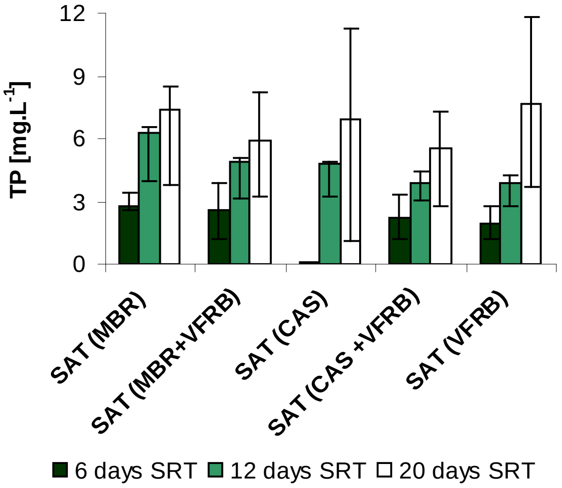

Figure 4-6 TP in the effluents at 6, 12 and 20 d SRT.

4.3.4 Solids and bacteria

All SAT demonstrated high removal of suspended solids with undetectable levels of TSS and turbidity below 1 NTU (data not shown). 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}.

The total *Coliforms* count remained below 2 CFU.mL^{-1} in SAT(MBR) at all SRTs (Figure 4-7). SAT(CAS) and SAT(CAS+VFRB) also provided low *Coliforms* count, respectively below 1 and 5 CFU.mL^{-1} at 12 and 20 d SRT, with 43 and 108 CFU.mL^{-1} at 6 d SRT. It is noticeable that SAT(VFRB) and SAT(MBR+VFRB) demonstrated an increase in total *Coliforms* count over time respectively with 28, 92 and 183 CFU.mL^{-1} and 1, 4 and 13 CFU.mL^{-1} for the three successive periods and the two SAT effluents. In addition, the *E. Coli* count ranged only between 0 and 3 CFU.mL^{-1} in all SAT effluents (data not shown). *Coliforms* removal in SAT, with the exception of SAT(MBR) where the feed count is very low, ranged between 54.70 and 99.97 %. Only SAT(CAS) provided constant removal above 99.77 % at all SRT in accord with the literature where more than 99 % removal was reported in full scale SAT or longer soil columns treating raw or secondary effluent (Crites, 1985;

Guessab *et al.*, 1993; Brissaud *et al.*, 1999; Gold, 1999; Jimenez and Chávez, 2004).

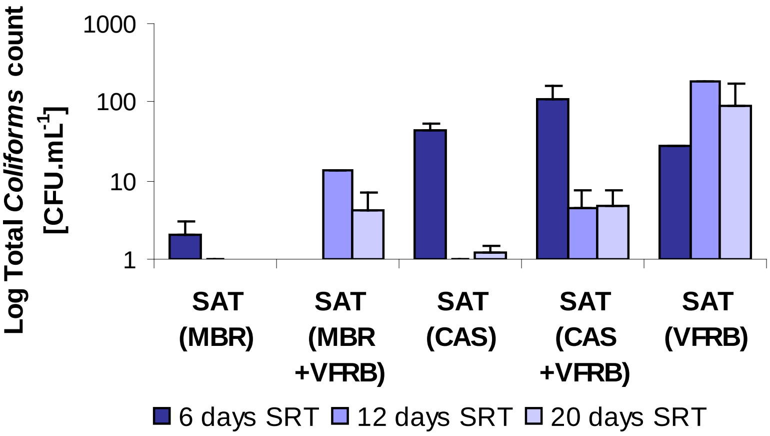

Figure 4-7 Total *Coliforms* count in SAT effluents at logarithm scale.

4.4 Impact of wastewater on SAT soil

4.4.1 Infiltration rate

Infiltration rate in SAT(MBR), with a mean of 34.7 cm.d^{-1}, was 2.2 to 2.5 times higher than in the other SAT columns at all times (Figure 4-8). Infiltration rate in SAT(MBR) was very variable for the first 6 months, ranging between 1.6 and 110 cm.d^{-1}. A significant reduction was observed after one 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 in SAT(MBR+VFRB) and SAT(CAS+VFRB) with means of 23.2, 17.8 and 4.6 cm.d^{-1} at 6, 20 and 12 d SRT. A different pattern occurred in SAT(VFRB) and SAT(CAS) where an increase in infiltration rate took place after 6 months before it was reduced again. In SAT(VFRB), infiltration rate increased from 16.3 to 30.0 cm.d^{-1} before it decreased to 3.4 cm.d^{-1} in the last period. SAT(CAS) provided the lowest infiltration rate at 6 d SRT with 2.7

cm.d^{-1} before it increased to 33.7 cm.d^{-1} after 6 months. For the last period infiltration rate in SAT(CAS) was higher than the first period with 8.4 cm.d^{-1}.

Infiltration rates at 12 d SRT in all columns, after one year of operation, and in SAT(CAS) at 6 d SRT were lower than the range observed by Lin *et al.* (2003). In the SAT where the soil was extracted for this study, rates ranged between 15 and 130 cm.d^{-1}. However, in the mentioned study (Lin *et al.*, 2003), the flooding periods were only 1 to 2 d with 5 to 7 d drying periods and the pre-treatment, an activated sludge process, included nitrification and denitrification. These differences presumably explain the discussed discrepancy in infiltration rates.

Variation of infiltration rate in SAT(CAS) and SAT(VFRB) may be related to temperature variation, successively of 7.8, 12.2 and 5.4 °C for the 6, 20 and 12 d SRT periods respectively. Lin *et al.*, (2003), 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 rate between the different SATs can be attributed to solids content and type in the influent. SAT(MBR) received no solids and started with the highest infiltration rate, followed in ascending order by SAT fed with tertiary and secondary VFRB and finally SAT(CAS) fed with the maximum solids content. Indeed, TSS is one of the main physical factors affecting SAT permeability (Loffler, 1969; Okubo and Matsumoto, 1983; Siegrist and Boyle, 1987).

Biological clogging is predominant in systems with prolonged periods of inundation like in this study (Rice, 1974). Despite SAT(CAS) and SAT(VFRB) being fed with a higher nutrient content, especially organics, than the other SAT columns, aerobic activity in these soils were similar to the other SATs (data not shown). This suggests that low infiltration rate and variations in these two SAT was caused by clogging via facultative anaerobic bacteria (Seki *et al.*, 1996; Shaw *et al.*, 1985; Okubo and Matsumoto, 1983; Platzer and Mauch, 1997) or extracellular polymeric substances (Vandevivere and Baveye, 1992; Nevo and Mitchell, 1967), which are temperature sensitive and controlled.

Despite the long resting periods used in this study, reduction of infiltration rate could be of concern in all SAT except SAT(CAS). Indeed, deep clogging phenomenon affects infiltration rate over the long-term and may require larger land areas to compensate, since there is no possibility of backwashing and drying like in direct injection wells or surface clogging in SAT basins (Fox, 1999).

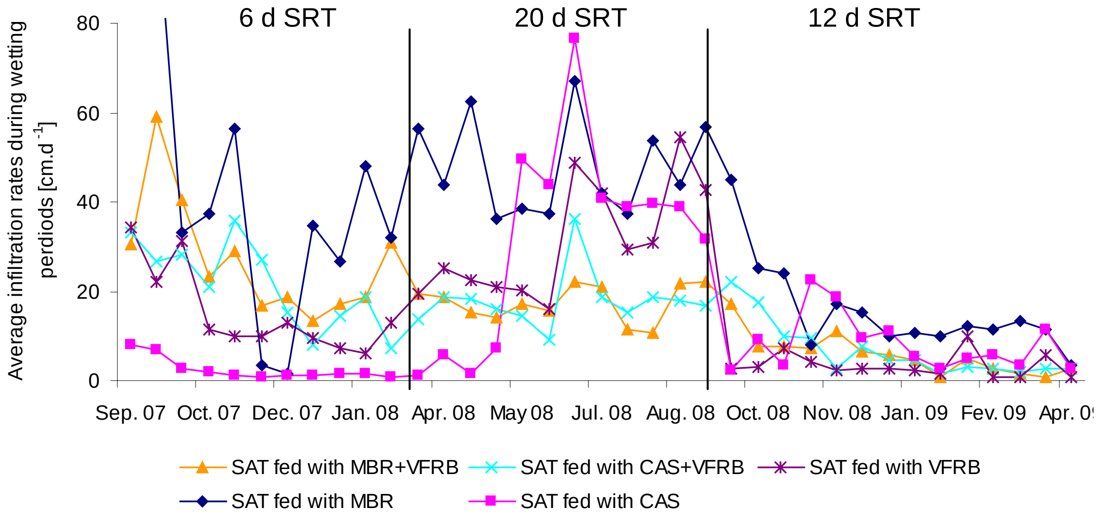

Figure 4-8 Infiltration rate in SAT soil fed with treated wastewater.

4.4.2 Changes in soil composition

The ratio of Oxygen to Carbon to Silica (O:C:Si) at the beginning of the experiment and for most of the column soils ranged between 2.5:0.9:1 and 3.0:2.0:1 except for SAT(MBR) and SAT(CAS+VFRB) at 20 d SRT with ratios of 2.8:0.8:1 and 3.0:2.6:1 respectively (Table 4-2). Nitrogen was present at about 6 % in the soil before the experiment started and was absent in all soils after flooding. An increase in aluminium was observed in all columns at 6 d SRT to double the initial percentage before flooding in SAT(MBR), SAT(CAS+VFRB) and SAT(VFRB). At 20 d SRT, the proportion of aluminium continued to increase for all SAT except SAT(CAS+VFRB) where a decrease was observed from 2.0 to 0.8 %. At 12 d SRT the aluminium ratio was similar to the beginning of the experiment. The elements S, P, Ti, Cl and Mn were absent or below 0.1 % in all soils. The element Ca ranged between 1.5 and 2.5 % before flooding and increased at 6 d SRT in SAT(CAS), SAT(CAS+VFRB) and SAT(VFRB) up to 7 % and 20 d SRT in SAT(MBR) and SAT(MBR+VFRB) above 7 %. At 12 d SRT, Ca ratio to the total of elements was about 3 % except for SAT(VFRB) where the proportion was lower at 2 %. The elements Na, K and Mg remained below 1 % except for K in SAT(MBR) with at 6 d SRT with 2.2 %. All SAT soils contained about 1 % Fe except SAT(MBR) and SAT(MBR+VFRB) at 20 d SRT where concentrations were 1.9 and 1.7 %.

The changes in proportions of most of the soil elements remained moderate. However, even minor changes may affect the soil quality in long-term effluent recharge. The variations in Ca can be related to the variations in pH and P removal. The increase in pH during the first period can be explained by the formation of dicalcium phosphate precipitate or similar basic forms (Cole *et al.*, 1953). The reduction in C percentage in SAT(MBR) at 20 d SRT may indicate carbonate dissolution, which causes over long periods a decrease in adsorption capacity of metals and phosphorus (Lin *et al.*, 2004). In the same way, a reduction in carbon content in SAT(CAS) soil at 6 d SRT can be explained by the previously mentioned hypothesis that denitrifying bacteria used the carbon from the soil as substrate. It is noticeable that SAT(CAS) did not demonstrate significant changes in P and N content compared to the other soils at 6 d SRT despite high removal of these elements being observed.

Table 4-2 Percentage weight of elements from SAT soil before and after feeding with wastewater effluents (at 6, 12 and 20 d SRT).

SRT [d]		6					12					20				
Element Weight [%]	Start	MBR	VFRB (MBR)	CAS	VFRB (CAS)	VFRB (primary)	MBR	VFRB (MBR)	CAS	VFRB (CAS)	VFRB (primary)	MBR	VFRB (MBR)	CAS	VFRB (CAS)	VFRB (primary)
C	25.0	27.4	30.3	20.2	25.7	23.0	26.3	29.4	24.4	28.8	25.2	15.5	21.8	20.4	37.4	26.4
O	47.5	47.6	48.6	50.6	48.4	50.0	48.5	47.1	50.3	48.1	49.5	52.0	49.8	50.3	43.4	49.8
Na	0.0	0.3	0.2	0.3	0.5	0.3	0.2	0.2	0.2	0.2	0.1	0.6	0.3	0.2	0.2	0.2
Mg	0.1	0.1	0.2	0.2	0.1	0.3	0.1	0.1	0.2	0.2	0.1	0.3	0.2	0.2	0.1	0.1
Al	1.1	2.6	0.9	1.7	2.0	2.0	1.4	1.1	1.3	1.2	0.9	2.8	2.4	1.9	0.8	1.7
Si	17.4	16.3	17.2	20.0	15.3	15.2	18.9	17.9	19.5	17.3	21.4	18.6	15.6	19.6	14.4	16.8
P	0.0	0.0	0.1	0.0	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.1	0.0
S	0.0	0.1	0.0	0.0	0.1	0.1	0.1	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.1	0.0
K	0.1	2.2	0.2	0.6	0.5	0.4	0.2	0.2	0.3	0.2	0.1	0.6	0.6	0.8	0.1	0.5
Ca	2.0	2.4	1.6	4.8	5.9	6.8	3.1	3.2	2.9	3.1	1.6	7.4	7.1	4.8	2.6	3.6
Fe	0.9	0.9	0.7	1.3	1.3	1.5	1.1	0.8	1.0	0.8	0.9	1.9	1.7	1.4	0.8	0.9

4.5 Soil aquifer treatment and the level of treatment required for safe and sustainable groundwater recharge

The different profiles of technologies considering treatment performance were generally retrieved in SAT performance (Table 4-3). The MBR provided bacteria free effluent and SAT(MBR) effluent had also the best bacterial quality. In the same way, SAT fed with VFRB effluent offered constant quality with enhanced nutrients removal. However, SAT(CAS) at low SRT demonstrated high removal of N and P despite CAS performances being moderate. In terms of SAT operation, the average infiltration was concordant with the solids content in SAT feed with the highest rate using MBR effluent and the lowest rates using CAS and VFRB(primary) effluents. Controversially, it seems the lower the infiltration rate the lower the clogging propensity of SAT soil but the higher impact of temperature variation on this rate.

Table 4-3 Main differences between treatments in regards to SAT performance and operation.

Pre-Treatment	MBR	CAS	Tertiary VFRBs	Secondary VFRB
Main features	Micro-filtration effluent free from solids and bacteria	Flocculation, high nutrient and particulate loading in the effluent	Sand filtration with low solids and nutrients effluent content	Sand filtration with high nutrient effluent content
Major impact on SAT performance	Best bacteria effluent quality	High N and P removal at low SRT	Enhanced and constant removal of N compounds and organics	Almost similar to tertiary VFRB but lower bacteria removal
Major impact on SAT soil	High infiltration rate but with apparent long term clogging	Very low infiltration rate with reversible clogging but with a high impact of temperature	Intermediate infiltration rate with moderate long term clogging	Low infiltration rate with a high impact of temperature

SAT produced an increased quality effluent relative to the study of pre-treatment based on reuse standards (Figure 4-9). Indeed, SAT as a unit provide a reduction in pollutants content of 5, 12 and 10 % respectively for 6, 12 and 20 d SRT. Improvements were greater for the effluents that were the least compliant towards reuse standards before SAT, to a maximum of 20 % increase in compliance for SAT(MBR) at 20 d SRT. However, it is likely variations in SAT effluent quality were not only a response to change in pre-treatment effluent quality and SRT but were also due to SAT soil evolution over time. Soil sorption capacity, infiltration rate, and other external conditions like temperature are all known to shift with time (Baveye *et al.*, 1998).

Independently of SRT, SAT improved COD content, and total *Coliforms* removal reached up to 100 % compliance with reuse standards. For example, CAS effluent at 6 d SRT demonstrated no compliance for *Coliforms* while SAT(CAS) obtained 78 % compliance after only one metre of soil passage. SAT(CAS) also demonstrated up to 98 % TP removal and 66 % TN removal at 6 d SRT. Indeed, the low SRT in the CAS allowed the simultaneous presence of nitrate and ammonium suggesting the potential for the ANAMMOX process (Fox *et al.*, 2006). However, the reduction in carbon content in the soil could indicate denitrification using the soil carbon as substrate. The high TP removal was also presumably influenced by the slow infiltration rate as the mineralization and fixing of phosphate by precipitation are slow processes (Lin and Banin, 2006). Other factors are high pH increasing the bioavailability of phosphorus as well as the amount of exchangeable Ca^{2+} that can precipitate P as a Ca-phosphate (Pereira and De Faria, 1998; Rupa *et al.,* 2001). This explains the release of P when changing the SRT from 6 to 20 d SRT, as the pH decreased (Barrow, 2002).

TN content was also reduced in SAT(CAS) and SAT(MBR) at 12 d SRT and in all effluents at 20 d SRT but only complied with restricted irrigation rather than SAT standards (Figure 4-9). Further nitrification took place in SAT degrading the compliance for nitrates but reducing the ammonium content of the CAS and VFRB(CAS) pre-treated streams at all SRTs and VFRB(primary). SAT increased pH at all SRTs and conductivity at 12 and 20 d SRT for all processes. The pH was increased above 8 in SAT(CAS) at 6 d SRT, hence while it was complying towards SAT reuse it failed to achieve irrigation reuse standards. The potential of SAT effluent reuse for restricted irrigation was high for SAT(MBR) and SAT(MBR+VFRB) at 6 and 20 d SRT, while SAT(CAS) and SAT(tertiary VFRB) were more appropriate options at 12 d SRT (Figure 4-9). When looking at the variation of compliance towards irrigation standards with time, SAT(MBR) effluent quality was degraded, while SAT(CAS) effluent quality was improved.

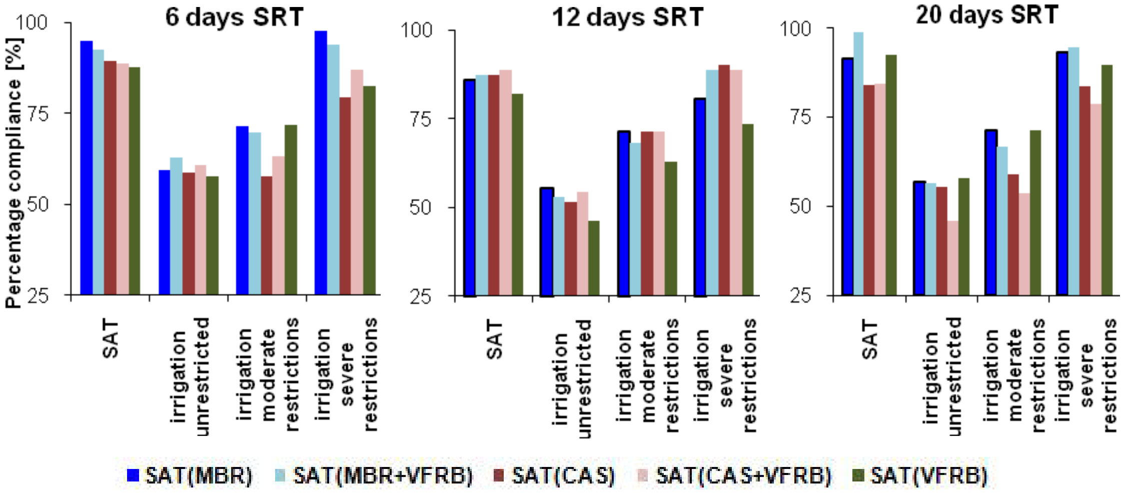

Figure 4-9 Compliance towards SAT and irrigation reuse standards (Bixio and Wintgens, 2006; EC, 2006; WHO, 2006; WHO, 2003; Kretschmer *et al.*, 2002; Ayers and Westcot, 1985, Pescod, 1992; Asano and Levine, 1998; Rice, 1974) after one meter of SAT, for the studied parameters.

4.6 Conclusions

SAT demonstrated high removals of solids and bacteria after only one metre of soil passage. SAT fed with CAS set up at 6 d SRT presented high removal of TN and almost complete removal of TP, but this was also associated with low infiltration rate, low temperature and high pH. SAT performance was not be correlated with SRT. However, it is possible temperature variation overwhelmed any such relationships. While SAT(CAS) effluent quality was increased over the 18 months of operation, SAT(MBR) effluent quality degraded over the same time. Infiltration rate in SAT(CAS) and SAT(VFRB) was affected by temperature. All SAT except SAT(CAS) underwent a reduction in infiltration rate over 18 months. The soil elemental composition showed moderate changes over time. In terms of compliance toward SAT reuse standards, little links could be found between the compliance of pre-treatment effluents and the compliance of these same effluents streams after 1 m SAT soil passage. From the most to the least complying effluents, treatment trains ranked this way: SAT(MBR 20d SRT+VFRB), SAT(MBR 6d SRT), SAT(CAS), SAT(CAS+VFRB) and SAT(VFRB). Compliance was similar for the last three treatment trains. In the case of an aquifer for irrigation purpose, SAT(MBR) and SAT(MBR+VFRB) were more compliant with irrigation reuse standards than the other treatment trains.

4.7 References

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5 CHARACTERISATION OF PHOSPHORUS REMOVAL IN SOIL AQUIFER TREATMENT AND EFFECT OF PRE-TREATMENT

5.1 Introduction

Phosphorus is an important compound for plant and biomass growth, however in excess it causes eutrophication in surface water leading to fish intoxication, development of anoxic zones and algal growth (Schindler, 1977). In agricultural applications, phosphorus can accumulate in soil and large quantities can be leached out into water bodies (Bennett *et al.*, 2001). For water in aquifers instead of surface water the problem is only delayed, as accumulated P will cause issues when reusing the recharged groundwater for irrigation, during drinking water treatment or natural discharge into surface water.

Soil aquifer treatment (SAT) is known to provide effective phosphorus removal by sorption and precipitation (Pescod, 1992; Lin *et al.*, 2006). However, several studies have reported release of phosphorus, locally high mobility and desorption (Walter *et al.*, 1995; Muneer and Lawrence, 2004; Sims *et al*., 1998). In addition to the characteristics of the soil and the operating conditions of the SAT, there are numerous factors affecting phosphorus sorption and precipitation, such as phosphorus loading, hydraulic loading rate, metals, temperature, organic matter, biosolids, particle size, reaction time, dissolved oxygen and pH (WEF, 1998; Garcia *et al.*, 2004; Shilton *et al.*, 2006; Mann, 1990).

This paper aims to understand how SAT pre-treatments affect phosphate sorption. The study of SAT columns fed with various treatment trains over 18 months showed important changes in phosphate removal over time, as temperature, type of pre-treatment along with significant pH and infiltration rate all varied (chapter 4). In addition, this study determines how and at which extent short term study of soil column and laboratory isotherms and kinetics experiments using phosphate solutions and treated wastewater effluents spiked with phosphate can be connected to long-term and field studies.

Isotherm adsorption models

The most relevant models describing sorption processes for phosphorus adsorption have been developed by Freundlich, Langmuir and Temkin (Freundlich, 1906; Langmuir, 1916; Temkin, 1941). These sorption models predict theoretical phosphorus retardation and maximum sorption capacity of the soil under specific conditions.

In the original equation, Freundlich proposed an empirical expression to the isothermal variation of adsorption of gas by the solid adsorbent with pressure that will be later applied to solutes (Equation 5-1).

$$\log\left(\frac{x}{m}\right) = \log(k) + \left(\frac{1}{n}\log C_e\right)$$

Equation 5-1

Where x is the mass of phosphate adsorbed [mgP], m the mass of adsorbent [g], k the Freundlich adsorption constant measuring the number of sites involved in sorption and related to the adsorption capacity [$mgP.g^{-1}$], n an empirical constant in Freundlich equation providing an estimation of the sorption intensity or strength and is above 1, and C_e the phosphate concentration at the equilibrium [$mgP.L^{-1}$].

However the Freundlich isotherm cannot predict correctly adsorption at higher pressures, beyond saturation, or multi-layer adsorption. Langmuir proposed a derived model to explain the variation of adsorption with pressure by including a relationship between the number of active sites and the surface (Equation 5-2). His assumptions included that there is a fixed number and homogeneity in size and shape of the adsorption sites on the solids surface, that each site can retain one molecule of gas, a constant amount energy is released during the process and a dynamic equilibrium exists between free and adsorbed molecules.

$$\frac{C_e}{q_e} = \frac{C_e}{q_{\max}} + \frac{1}{b_1 q_{\max}}$$ **Equation 5-2**

Here q_e the mass of solute adsorbed per unit mass of adsorbent [mgP.g^{-1}], q_{max} the monolayer capacity or total capacity of adsorbent for adsorbate [mgP.g^{-1}] and b_1 the Langmuir constant related to the heat of adsorption.

However, the Langmuir model is limited to low pressure conditions and has many of the same limitations as Freundlich's model. Temkin's model is also used to describe sorption onto a solid and is based on the postulate that the free energy of adsorption increases linearly with the coverage because of the heterogeneity of the surface or the lateral interactions (Equation 5-3). It is limited to specific pressures in regions of high coverage (Asnin *et al.*, 2001).

$$\exp(q_e) = \exp(K_1) + K_2 C_e$$ **Equation 5-3**

K_1 the Temkin constant [mgP.g^{-1}] and K_2 the second Temkin constant related to the energy of adsorption [L.mgP^{-1}].

5.2 Materials and Methods

The studied soil was provided courtesy of the Shafdan SAT site, Israel, and was the same soil used for the long term soil columns operated under 7/7 d rotations of drying and wetting periods (Chapter 4). Sorption isotherms were obtained following the method from Rowell, (1994). The shaking time was however set up at 3 d instead of 24 h as SAT soil was expected to have a large adsorption capacity (Lin and Banin, 2006). A 7 d shaking experiment was also carried out and no significant difference could be found with the 3 d experiment. Additional sorption isotherms were acquired by replacing the deionised (DI) water with the pilot plant effluents. Experiments were carried out in triplicate.

The effluents used in the laboratory experiments were taken from treatment trains including a membrane bioreactor (MBR), a conventional activated sludge (CAS) and three vertical flow reed beds (VFRBs) fed with MBR, CAS and primary effluent (Chapter 3). MBR and CAS were operated at 8 h HRT and 12 d

SRT. The data were then fitted to Freundlich, Langmuir and Temkin's models according to the linearised equations 5-1, 5-2 and 5-3.

Bed volume experiments were carried out using soil column of 2.6 cm internal diameter filled with 20 cm of sand packed at 1.5 $g.cm^{-3}$ density (Figure 5-1). The column was operated for 7 d under saturated conditions with a constant head of 5 cm and fed with a phosphate solution of 8 $mgP.L^{-1}$. Samples were taken every 1 to 3 h until 80 % breakthrough and then every 6, 12 and 24 h. Regeneration studies on constructed wetland are sometimes undertaken to determine the reusability of the adsorbent following rinsing with water (Frazer-Williams, 2007). However, in this study the length of drying and wetting periods were chosen to give an insight of sorption sites regeneration during long-term SAT column operation. In addition, this experiment was carried out to highlight and isolate the specific effects of phosphate on soil at representative SAT operating conditions.

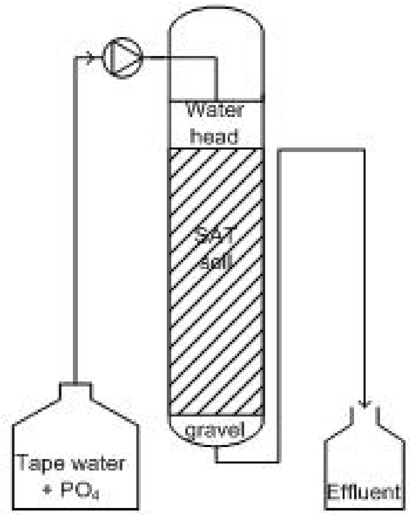

Figure 5-1 Column set up for bed volume experiment.

The resulting curve was fitted to the model described by Yoon and Nelson, (1984), using the equation 5-4. The experiment was repeated three times allowing 7 d of resting in between. It is noticeable that pH increased slightly after soil passage but remained constant in all BV experiments. Values were between 7.0 and 7.5 in the effluent compared to 6.8 in the influent.

$$\ln\left(\frac{C_t}{C_i - C_t}\right) = K_{YN}(t-\theta)$$ **Equation 5-4**

Where C_t is the phosphate concentration [mgP.L^{-1}] at time t [h], C_i the initial or feeding phosphate concentration [mgP.L^{-1}], K_{YN} the rate constant [h^{-1}] and θ the breakthrough time when $C_t = 0.5\ C_i$ [h].

Kinetics experiments were carried out using DI water spiked with phosphate solution to obtain a final concentration of 9.4 mgP.L^{-1}. Calcium chloride, 10 mM, was added to maintain soil stability. For each Erlenmeyer flask, 11 samples of 2 g of soil in phosphate solution was shaken at 180 rpm and filtered after 0, 0.25, 0.5, 1.5, 6, 12, 24, 36, 48, 60 and 168 h. The experiment was repeated three times. Data were fitted to zero, first and second order kinetics, diffusion model, modified Freundlich and Elovich equations 5-5, 5-6, 5-7, 5-8, 5-9 and 5-10 (Griffin and Jurinak, 1974; Cooke, 1966; Kuo and Lotse, 1974; Chien and Clayton, 1980).

$$C_t = -k_0 t + C_0$$ **Equation 5-5**

$$\ln C_t - \ln C_0 = k_1 t$$ **Equation 5-6**

$$\frac{1}{C_t} - \frac{1}{C_0} = -k_2 t$$ **Equation 5-7**

$$q_e = r t^{0.5} + b$$ **Equation 5-8**

$$\ln q_e = \ln k(C_0) + \frac{1}{n}\ln t$$ **Equation 5-9**

$$q_e = \frac{1}{\beta}\ln(\alpha\beta) + \frac{1}{\beta}\ln t$$ **Equation 5-10**

With C_t the phosphate concentration [mgP.L^{-1}] at the time t [h] and k_0, k_1 and k_2 the constants of zero, first and second order kinetics [mgP.L^{-1}.h^{-1}], [h^{-1}], [L.mgP^{-1}.h^{-1}], r [mgP.h$^{0.5}$.g^{-1}], b [mgP.g^{-1}], k rate constant [h^{-1}], n, α and β [g.mgP^{-1}].

Phosphate content was determined using spectrophotometry (Spectroquant cell tests, Merck, VWR International, Poole, UK). The pH and conductivity were measured using a conductivity/pH meter (Jenway 3540, Bibby Scientific Ltd

T/As Jenway, Essex, UK). Coefficient of determination (R^2) and standard error on regression (σ) were used to define the fitting on the different models equations. R^2 indicates the proportion of the variance in the dependent variable that is predictable from the independent variable. The σ evaluates the accuracy of the regression by measuring the average amount that the regression equation over or under predicts the true value.

5.3 Sorption and precipitation of phosphorus in SAT soil

5.3.1 Sorption isotherms

The adsorption isotherms obtained with the pilot plant effluents showed main differences below 15 or above 40 mgP.L^{-1} equilibrium content as curves converge in this interval (Figure 5-2). The soils in VFRB(CAS) and VFRB(primary) solutions provide a better adsorption with lower equilibrium concentrations, while the soils in the other solutions offer maximal adsorption at high equilibrium concentrations. Freundlich equations gave a better estimation of the isotherm using DI water with coefficient of determination of 0.97 compared to 0.80 for Langmuir. However, all the other isotherms, obtained with treated wastewater, fitted the Langmuir equation better, with coefficients of determination between 0.88 and 0.99. In addition, the variance analysis of the Freundlich modelled data (using F test) demonstrated that the regression was not significant. This implies that saturation of the adsorbent surface occurred when using wastewater effluent instead of DI water. This suggests the release of phosphate from the decomposition of organic matter complexes as observed by Guppy *et al.*, (2005).

The Temkin model was not the best estimation of the isotherms, but it offered the lowest standard error of the estimate with 8 and 9 % for the isotherms using CAS and MBR effluents compared with an error of 21 and 13 % for the

Langmuir model. The higher accuracy of the Temkin model is most likely because it takes into account the heterogeneity of the surface on which the adsorption energy decreases with increasing coverage.

Considering the Freundlich analysis, SAT soil in DI water had the lowest capacity of adsorption a with K value of 0.132 mgP.g^{-1} (Table 5-1). The adsorption capacity of the soil in DI water was below all soils shaken in wastewater effluent, despite the fact that competition with other ions in the effluent was expected to reduce the adsorption capacity (Lin and Banin, 2006). This is probably explained by the interaction with wastewater solutes and particles favouring or stabilising phosphate adsorption relative to DI water. However at high equilibrium concentration, a different trend applied: soils in DI water presented higher sorption intensity with n of 1.923 compared to most of the soils in treated wastewater.

When considering isotherms in wastewater effluent, soil in CAS effluent had the highest adsorption capacity with q_{max} of 0.503 mgP.g^{-1} followed by soils in MBR and VFRB(MBR) with 0.431 and 0.417 mgP.g^{-1}, VFRB(primary) with 0.344 mgP.g^{-1} and VFRB(CAS) with 0.318 mgP.g^{-1} (Table 5-1).

Previous studies on aquifer soils fitted P adsorption to Langmuir's model with variable sorption capacities (Table 5-1). In this study, the adsorption capacity in DI water, with 0.132 mgP.g^{-1}, was in accordance with Lin and Banin, (2006), who found an adsorption capacity of 0.110 mgP.g^{-1} using a similar soil. They also observed that kinetic limitations led to an underestimation of P adsorption and this has been verified in this study. Kulabako *et al.*, (2008), found a higher adsorption capacity at 0.600 mgP.g^{-1} close to the capacity found with CAS effluent of 0.503 mgP.g^{-1} in this study. Both the mentioned studies proposed that P sorption and mobility were related to concentration of Ca, Fe, Al, Mn and carbonate at the soil surface, and organic carbon content of the soils.

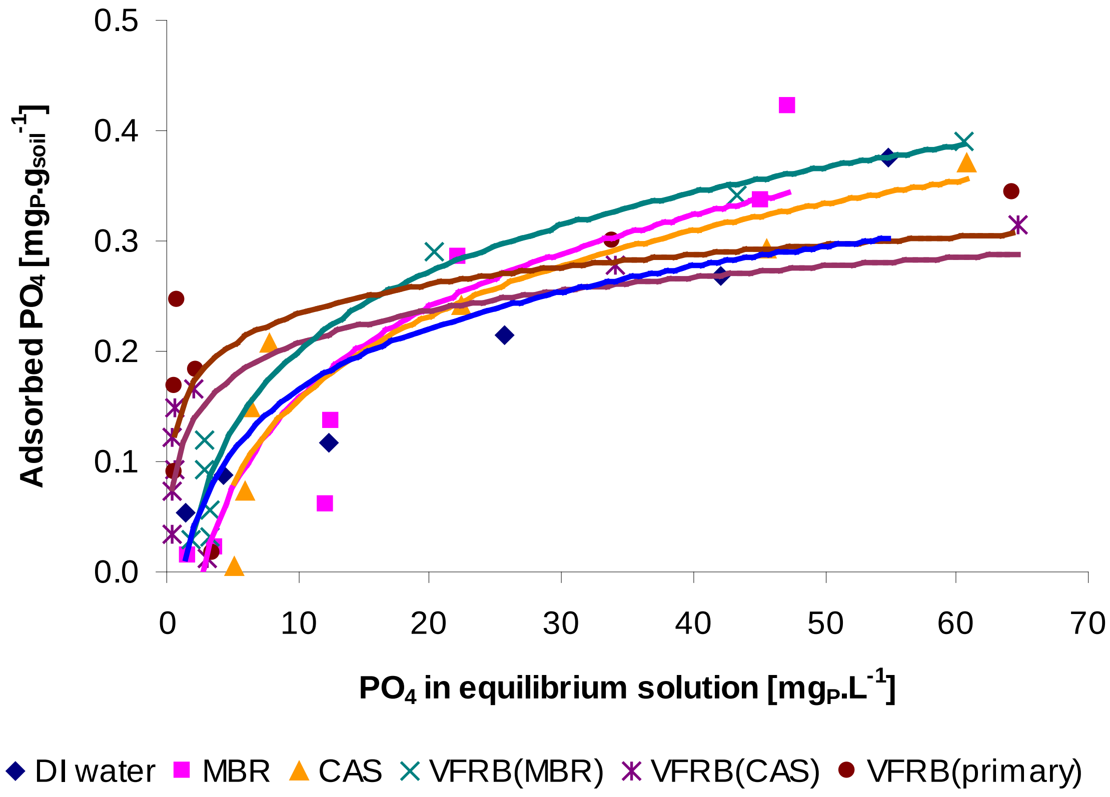

Figure 5-2 Isotherms of adsorption with different solutions.

Table 5-1 Maximum Langmuir adsorption capacity compared with literature values.

Soil type	**Langmuir q_{max} [mgP.g-^{1}]**	**reference**
calcareous sand stones (SAT) (in DI water)	0.132	This study
calcareous sand stones (SAT) (in MBR effluent)	0.431	
calcareous sand stones (SAT) (in CAS effluent)	0.503	
calcareous sand stones (SAT) (in VFRB(MBR)	0.417	
calcareous sand stones (SAT) (in VFRB(CAS)	0.318	
calcareous sand stones (SAT) (in VFRB(primary)	0.344	
clay sand (urban area)	0,600	Kulabako *et al.* (2007)
calcareous sand stones (SAT)	0,110	Lin and Banin (2006)
silt and clay (mesic organosol) +limestone	1,042	Andrade *et al.* (2002)
silt, clay and sand (melanic gleysol) +limestone	5,014	
sand (haplic gleysol) +limestone	0,548	
silt and clay (fluvic neosol) +limestone	0,728	
limestone, cinder and loess (constructed wetland)	2,002	Guan *et al.* (2009)
clay and limestone	1,200	Gray *et al.* (2000)

5.3.2 Bed Volume

Bed volume trials were carried out in order to confirm the P sorption capacity obtained from isotherm studies and to evaluate the life expectancy of the SAT soil surface. The shape of the breakthrough curve clearly exhibited a mass transfer zone between 4 and 15 BVs (Figure 5-3). The phosphate content in the column effluent remained below 0.05 mgP.L^{-1} (drinking water standard) for the first 3 BVs and below 2 mgP.L^{-1} (SAT reuse standard) until 9 BVs. The sorption capacity, as deduced from the curve, was 0.041 mgP.g$_{soil}^{-1}$. The sorption capacity was three times less than the one obtained from isotherm tests. However, the difference in contact time needs to be addressed with a hydraulic retention time of 1.7 h in the column compared to 72 h for the isotherm experimentation. From fitting the data to Yoon and Nelson's model, the breakthrough time (θ) was 11.84 h and the rate constant K_{YN} 0.47 h^{-1}. A discrepancy between the real and modelled data occurred between approximately 8 and 25 BVs, this might be an indicate pore diffusion control being dominant over external film control (McCabe *et al.*, 1985).

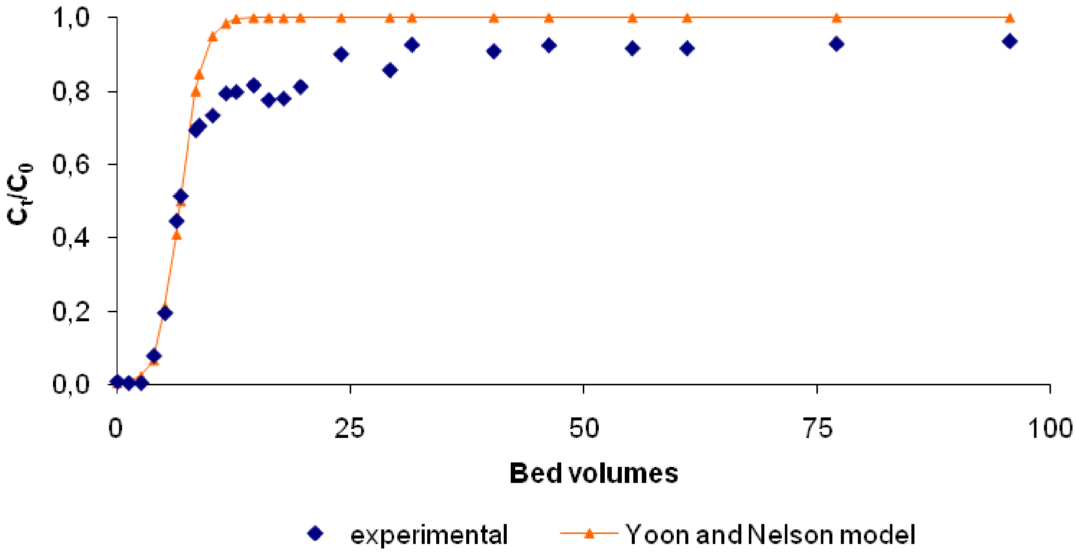

Figure 5-3 Elution of phosphate with bed volumes and the fitting to Yoon and Nelson's model.

5.3.3 Kinetics of sorption

Three phases were observed when plotting the phosphate sorption with reaction time (Figure 5-4-A-). One fifth of the phosphate was removed within the first 30 min, which corresponded to 40 % of the total removal after 3 d. Then up to 24 h, the phosphorus sorption rate slowed down and even desorption was observed. Finally, a steady slow rate of sorption was observed, with 25 % of the total removal in 47 h.

This shape has been described for phosphorus sorption kinetics by numerous authors (Hansen *et al.*, 1999; Cheung and Venkitachalam, 2006; Ryden *et al.*, 1977; Kato and Owa, 1989; Jones and Amador, 1992), though the lag phase was questioned by Barrow *et al.*, (2000). The fast initial rate was attributed to sorption onto mineral surface such as iron and aluminium oxides and precipitation with exchangeable calcium from the calcareous fraction of soil, while the slow rate suggested the saturation of sorption surface and diffusion into pores (Cheung and Venkitachalam, 2006; Ryden *et al.*, 1977).

The best fit of the sorption data was with first and second orders kinetics, with R^2 of 0.93 and 0.94 and σ of 0.08 and 0.01 respectively (Table 5-2, Figure 5-4-B-). This implies that the sorption rate is proportional to the soil content in solution or to the number of available sorption sites (Van Riemsdijk *et al.*, 1975; Novak and Petschauer, 1979). However, when considering the slow rate after 24 h, a good fit to the diffusion model was observed, with R^2 of 0.98 and σ of 0.002. This suggests the adsorbate content was reduced and the adsorbent surface saturated in accord with Cheung and Venkitachalam, (2006), (Figure 5-3-C-).

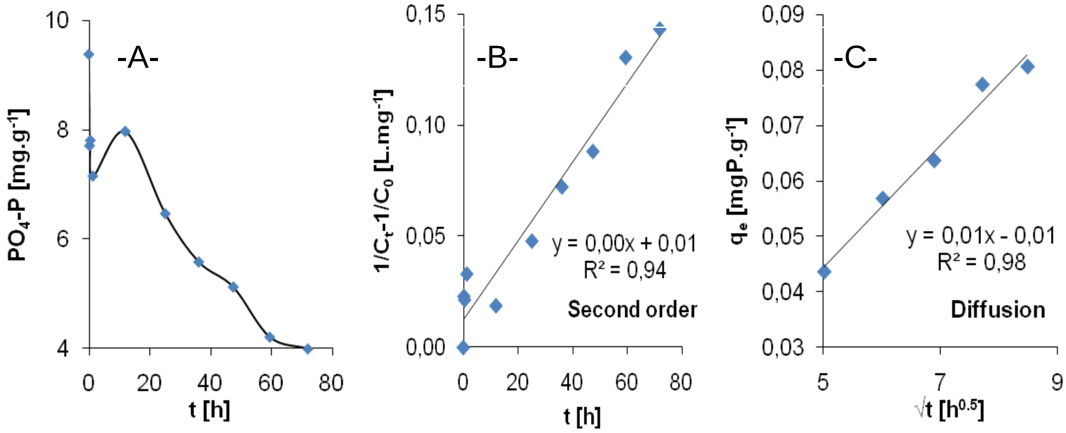

Figure 5-4 -A- Kinetics of phosphate adsorption and -B- fitting to second order kinetics and -C- fitting to diffusion model after 25 h.

Table 5-2 Coefficients of fitted kinetics models (R^2 coefficient of determination, σ standard error on regression).

	Kinetics order			Diffusion	Modified Freundlich	Elovich
Models	0	1	2			
Coefficients	k_0=0,068	k_1=-0,010	k_2=-0,002	r=0,008 b=0,009	n=5,7 k=-2,6	β=133 α=1,03
R^2	0,89	0,93	0,94	0,87	0,65	0,75
σ	0.63	0.08	0.01	0.01	0.33	0.01

5.3.4 Potential regeneration of SAT by long resting period

The first bed volume experiment was carried out using a preliminary saturation with tape water for 2 d followed by 7 d flooding with a phosphate spiked solution before draining the column and leaving it to rest for 7 d. However, for the next flooding, the phosphate solution was directly used for saturation and flooding. Consequently, aerobic condition was present at start for the second and third wetting periods in contrast with the first wetting period.

The infiltration during the second wetting period showed an unexpected shape for the first 10 BVs (Figure 5-5-B-). From the start 70 % of phosphate was eluted in less than 2 BVs suggesting that adsorbed phosphate from the previous flooding became desorbed when wetting the column again. Between 2 and 10 BVs, phosphate content in the effluent was stable at 50 % elution. The Yoon and Nelson shape, (1984), applied in the mass transfer zone between 10 and 16 BVs. Until 26 BVs, the infiltration rate was only of 0.36 mL.min^{-1} compared to 0.69 mL.min^{-1} during the first wetting period thought to be caused by pore blockage by precipitate or adsorbed phosphate from the previous flooding (Figure 5-5-B-). The infiltration rate increased exponentially after 26 BVs which corresponded to breakthrough of phosphate. Similarly to the first wetting period, a disparity to Yoon and Nelson's model was observed with a constant elution of 74 % of initial concentration between 16 and 26 BVs.

The third flooding period showed no similarity in shape with the two previous experiments (Figure 5-5-A-). The sorption sites seem to be regenerated and for the first 8 h only 7 % of injected phosphate was eluted. However the infiltration rate was very slow during this period at only 0.04 mL.min^{-1}, corresponding only to 0.24 BVs (Figure 5-5-B-). From 0.74 to 21 BVs, phosphate elution started rapidly before reaching a plateau at 30 % elution. During this phase, the infiltration rate increased to 0.30 mL.min^{-1} and remained constant until 50 BVs. Breakthrough occurred at 72 BVs while infiltration rate peaked at 0.96 mL.min^{-1}, followed by a reduction to 0.16 mL.min^{-1} at 84 BVs.

Breakthrough time seemed to be retarded after the successive cycles respectively at 12, 47 and 72 BVs for the first, second and third flooding periods. The maximum absorption capacity was only lower during the second experiment at 0.013 mgP.g^{-1} compared to 0.041 mgP.g^{-1} for the first and third wetting periods. This suggests a potential regeneration of the adsorption capacity by rotation with drying period.

When considering the effect of infiltration rate variations, the slope obtained by plotting log P adsorbed against log BV is relatively constant (Figure 5-5-C-). However, the y-intercept was 0.75, 0.15 and 0.20 for the first, second and third flooding periods respectively. As the intercept decreases, the infiltration rate increases in accord with Yoon and Nelson's findings, (1984). Variation of infiltration rate cannot be attributed with certainty to phosphate sorption as it is also affected by saturation processes and other effects of drainage and flooding hydraulics.

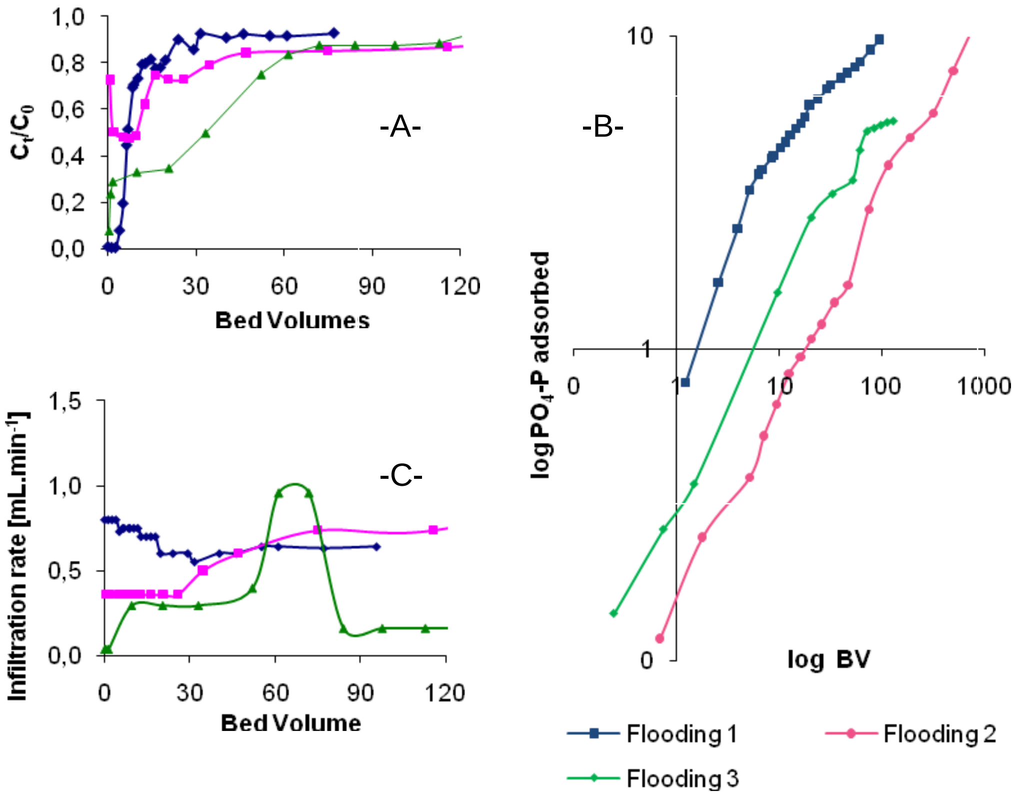

Figure 5-5 -A- Elution fraction, -B- variation of infiltration rate with bed volume and –C- logarithmic variations of adsorbed phosphate with bed volume.

5.4 Effect of effluent type on phosphate removal in soil aquifer treatment

For the first 6 months when CAS and MBR were set up at 6 d SRT, the SAT columns provided stable and consistent phosphate removal with mean values between 36 and 49 % and effluent content between 1.63 and 3.54 mgP.L^{-1} respectively for SAT(MBR) and SAT(VFRB) (Figure 5-6-A-). In SAT(CAS), a dissimilar pattern was observed with an effluent content below 0.10 mgP.L^{-1} corresponding to 98 % P removal. However at 12 and 20 d SRT, large breakthrough peaks appeared in all SAT and overall reduction in performance was observed. At the time, these variations were thought to be related to changes in pH and the infiltration rate (Figure 5-6-B- and -C-). Indeed, the pH in SAT fed with CAS set up at 6 d SRT was significantly higher with 8.33 compared to average of 7.80 in the other columns (Figure 5-7-B-). In the same way, the phosphate leakage after 6 months was accompanied by a decrease in pH. According to Brady and Weil, (1999), the major part of phosphate fixation is related to calcium phosphate formation in this range of pH and with this soil type. Indeed, the SAT soil was composed in majority of calcium carbonate with 24.6 % weight, while Al, Fe and Mn accounted only for 1.3 %. Consequently, the reduction of pH, that affected the main pathway for P removal namely calcium carbonate, reduced significantly the phosphate removal. Phosphate variations were also linked with variations in infiltration rate for SAT(CAS) as the infiltration rate was 20 folds the other SAT rates, but this also meant that less phosphate was applied to the soil (Figure 5-6-C-).

The maximum adsorption capacities were underestimated in the sorption isotherms and bed volume experiments when comparing with long term SAT columns. It was found that up to 0.80 $mgP.g^{-1}$ was achieved per wetting period during the first months in the SAT columns instead of 0.50 $mgP.g^{-1}$ determined with Langmuir model. This tallied with Lin and Banin, (2005), who observed higher sorption in a SAT column than in isotherms experiments due to kinetic limitations when comparing to the regeneration of sorption sites with the cycles of wetting and drying periods in SAT column.

In terms of infiltration rate, the first two bed volume experiments provided up to 10 times the infiltration rate of the pilot plant and 100 times the rate of SAT(CAS). The third bed volume experiment showed an infiltration rate closer to the pilot plant for the first 50 BVs which corresponded to a higher sorption capacity than the first two wetting periods. This suggests that the longer residence time in SAT(CAS) encouraged higher phosphate adsorption (Garcia *et al.*, 2005; Sakadevan and Bavor, 1999; Howard-Williams, 1985). In addition, the higher phosphate and organic loading in the other SAT caused by a higher infiltration rate is a feature of decreasing the P binding capacity of soil (Brauer, 2007; Mullins, 1991; Indiati, 2000).

The phosphate leakages observed in all SATs after 6 months were more difficult to explain. The drop in pH suggests an exhaustion of sorption onto calcium carbonate in the soil, the major adsorbent constituent of the studied soil. This is predicted to result in a shift in removal mechanism from complexation with calcium to fixation with metals that were in lesser proportion in the soil (Brady and Weil, 1999). In addition, the P release from all soils after 6 months of SAT operation can be attributed to soil equilibrium processes after the high P removal in SAT during the first months of operation as observed by Rosolen, (2000).

The estimated lifetime of SAT columns treating wastewater effluent ranged between 1 week and 2 years. These values were derived from the maximum adsorption capacity determined with BV experiment and the lowest and maximum P loading in SAT columns. Estimates were higher at between 1 month and 5 years using kinetics results of DI water tests. Considering SAT columns infiltration rate, phosphate influent content and q_{max} from kinetics in wastewater effluents, the lifetime estimation of SAT columns for phosphate retention ranged between 9 months for SAT(VFRB) and 18 years for SAT(CAS) set up at 6 d SRT (Table 5-3).

Considering the 5 to 30 m depth before the applied water reaches groundwater, average retardation factors ranged between 26 to 156 years comparable with the estimates of Lin and Banin, (2006). These authors observed up to 50 % phosphorus removal in field studies with long retardation factors between 15 and 235 years, though with shorter flooding periods than in this study. However, the retardation factor for SAT(CAS) at 6 d SRT ranged between 169 and 1014 years, possibly related to a slower infiltration rate than the other treatments.

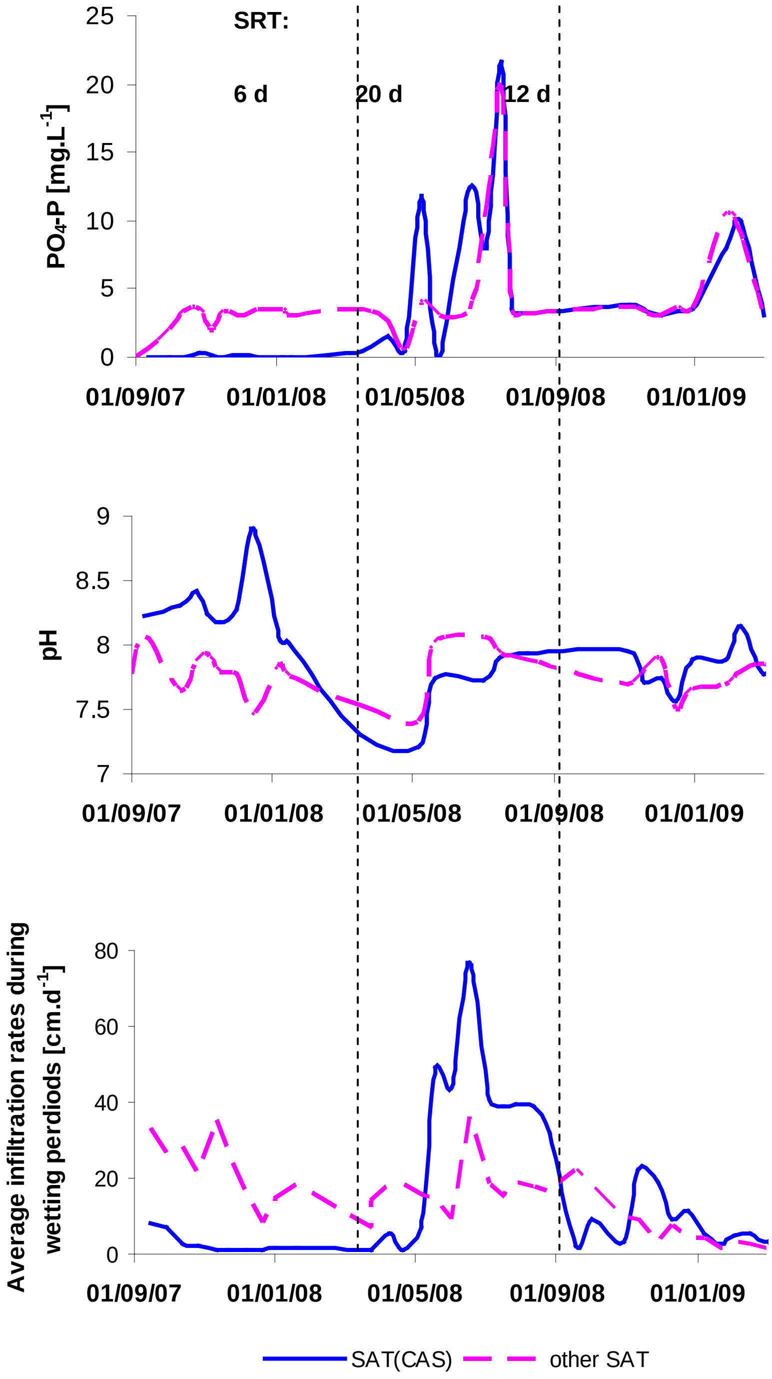

Figure 5-6 -A- Variations of phosphate content in SAT effluent with time -B-variation of pH and -C- variation of infiltration rate.

Table 5-3 SAT column lifetime for phosphate retention (m months; y years of flooding) using q_{max} determined in wastewater effluents, infiltration rate and phosphate effluent contents from SAT columns experiment.

SRT [d]	SAT(MBR)	SAT(CAS)	SAT(MBR+VFRB)	SAT(CAS+VFRB)	SAT(VFRB)
6	10 m	18 y	18 m	17 m	22 m
12	8 m	11 m	20 m	13 m	9 m
20	3 y	5 y	7 y	5 y	8 y

5.5 Conclusion

Isotherm experiments underestimated the adsorption capacity compared with data from long term study such as SAT columns. Nevertheless the ranking of sorption capacity observed with soil shaken in treated effluents was retrieved in the long-term experiment. The isotherm experiments using different types of solute demonstrated the inadequacy of using DI water as results fitted the Freundlich model, while isotherms using wastewater effluents fitted the Langmuir model most closely.

The bed volume experiment provided a lower adsorption capacity than isotherm or long-term soil column experiments, though the retention time in the bed volume study was also smaller. When approaching breakthrough of phosphate, a deviation from the Yoon and Nelson model was observed which suggested a change in mechanism in favour of diffusion over external film control. This was confirmed by the kinetic study, where second order kinetics was followed by a phase where the slow rate fitted the diffusion model. Soil regeneration during resting period highlighted the influence of saturation, aerobic conditions and infiltration rate on P sorption. Restoration of sorption capacity was observed over the three cycles of wetting and drying periods. However, variations in breakthrough shape were detected thus the longer-term study was necessary to resolve stabilised behaviour.

An increase in pH after soil passage indicated reactions between phosphorus and the calcium carbonate fraction of the soil. This confirmed the correlation between reduction in pH and the phosphate leakage after 6 months in the long-term soil columns caused by surface carbonate exhaustion.

The laboratory study confirmed the interactions observed in long term SAT columns between infiltration rate and P application and removal in both ways. The use of treatment trains such as CAS and VFRB(primary) leads to superficial SAT soil clogging because of the high solids and nutrients loading and hence leads to a low infiltration rate. In these treatment trains, higher P removal was observed without higher soil clogging. However, all treatment trains are limited by the soil adsorption capacity. Once the capacity reached, the perturbations of the soil, such as pH and soil carbon content, can have a cascade of adverse effects on general SAT operation and performances.

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6 METAL REMOVAL IN WASTEWATER TREATMENT TRAINS AND SOIL AQUIFER TREATMENT

6.1 Introduction

An excess of metals in freshwater has been identified as toxic and endocrine disrupting compounds for the aquatic environment (Lester, 1987; Brikett and Lester, 2003). In the case of wastewater reclamation for groundwater recharge, concerns have been raised regarding heavy metal accumulation in soil after wastewater infiltration (Bouwer and Chaney, 1974; Streck and Richter, 1997; Banin *et al.*, 2002). The risk of metal leakage, reduction of metal retention capacity and other various effects on soil stability and clogging have been demonstrated in the literature (Baveye *et al.*, 1998; WHO, 2004).

In some cases, natural metal content in groundwater can be lower than drinking water standards and the quality of the water to be infiltrated should be in accord with the targeted aquifer. For instance, zinc content in some groundwater has been reported below 40 ppb, some 100 times below the WHO drinking water standards (WHO, 2004). Similarly, for irrigation reuse purposes, risk of soil destabilisation and pollution, as well as toxicity for plants and accumulation in crops are the main concerns when applying water containing metals (Metcalf and Eddy, 2003).

For these reasons, the fate of metals in the wastewater flow sheet is critical to offset the risk of failure to comply with reuse standards and the epidemiological and environmental consequences. Most studies on metal removal focus on conventional activated sludge (CAS).Recently, with the expansion of membrane filtration, pilot plants including membrane bio-reactors (MBRs) have started to emerge (Innocenti *et al.,* 2002; Fatone *et al.*, 2005). CAS and MBR offer alternative options for metal removal by their intrinsic differences in design and operation. CAS has the advantage of providing consistent flocs, where metals can be adsorbed and removed via settlement. However, MBR presents the capacity for higher solids retention and to produce less or no sludge, reducing further treatments required for sludge reuse or disposal. Müller *et al.*, (1995), reported better removal with MBR than CAS for Cd, Cr and Ni and the opposite trend for Cu, K, Mg, Ag and Hg.

Correlation of metal removal with various parameters, such as organic content and pH, have been clearly established (Goldstone *et al.*, 1990a; 1990b; Chen and Gu, 2005), yet the effect of sludge retention time (SRT) is still contested (Stephenson and Lester, 1987; Innocenti *et al.*, 2002).

Meanwhile, there has been increased attention on tertiary treatments operated for metal removal. Constructed wetlands are often specified as an additional treatment step for metal removal where a specific matrix such as bauxite or metal hyper-accumulating plants are used (Wood and McAtamney, 2004; Prasad, 2003). As with a classic sand bed, removal is thought to occur by precipitation, filtration and biological removal, with micro-organisms present in soil and near the roots, and finally plant uptake also operative for constructed wetlands (Kirkham, 2006; Vymazal *et al.*, 2009). The final step is soil aquifer treatment (SAT), where removal mechanisms for metals involve adsorption, precipitation, biotransformation and immobilisation into soil, with removal ranging from 50 to 90 % depending on the metal, and the biological and chemical environment (Alloway, 1995; Vanbroekhoven *et al.*, 2007). Heavy metals are mainly retained within the first metre of the SAT soil by complexation and surface precipitation onto iron oxides and carbonate (Lin *et al.*, 2004). The main factors affecting metal removal are the sorption capacity of soil, the soil pH, the oxidation-reduction potential of the water, metals loading, concentration of organic matter, and to a lower extent, particle size in the water and flow rate (Kirkham, 2006; Lee *et al.*, 2004). The presence of ligands, such as hydroxides, clays, sulphides, natural organic carbon and ions, also affects metal immobilisation and transformation (Wang and Mulligan, 2006).

Despite the potential of SAT soil for metal removal, especially in carbonate and iron rich soils, issues and risks related to the sustainability of removal capacity and the release of retained metal have already been addressed (Magaritz *et al.*, 1990; Lin *et al.*, 2004; WHO, 2004). Margaritz *et al.*, (1990), observed enrichment in Zn and Cu in the mobile phase of a carbonate aquifer material after anthropogenic contamination. Similarly, Lin *et al.*, (2004), observed a decline in soil affinity for metal as the degradation of organic matter lead to a decrease in pH and consequently the dissolution of carbonate and Mn oxides. The WHO, (1997, 2004), reported rises in dissolved Al up to 90 ppm due to acidic environmental exposure, like acid rain water or mine drainage, compared to 1 to 50 ppb in neutral water. Conversely, in other studies the impact of wastewater or acid rain on soil pH was minimal and metal desorption or dissolution was not observed (Drewes and Jekel, 1996; Lee *et al.*, 2004).

This study aims to compare mechanisms and performance of metal removal by CAS and MBR and to assess the effect of SRT, with the final objective of SAT and irrigation reuse. This paper also assesses the potential benefits of secondary and tertiary VFRBs for metal removal after CAS or MBR, and for SAT pre-treatment as a soil buffering step. Furthermore, this paper will examine the effect of treatment technology and operation on sustainability of SAT metal removal.

6.2 Method

6.2.1 Pilot Plant

Experiments were carried out at Cranfield University with a treatment train involving a CAS followed by VFRB, a MBR followed by VFRB and a VFRB fed with primary effluent, all running in parallel. Each unit was followed by a SAT soil column (Figure 6-1).

CAS and MBR were run at 8 h HRT and successively 6, 20 and 12 d SRT. The MBR design included a tubular microfiltration membrane submerged in a 35 L aerated biomass tank, while the CAS was composed of a 30 L aerated biomass tank, a 2 L pre-anoxic zone and a 7 L clarifier. The MBR was set up in submerged membrane configuration, with a 50 cm long membrane module composed of 21 tubes of 0.08 μm pore size, and 6 mm lumen diameter, corresponding to 0.2 m^2 membrane surface area.

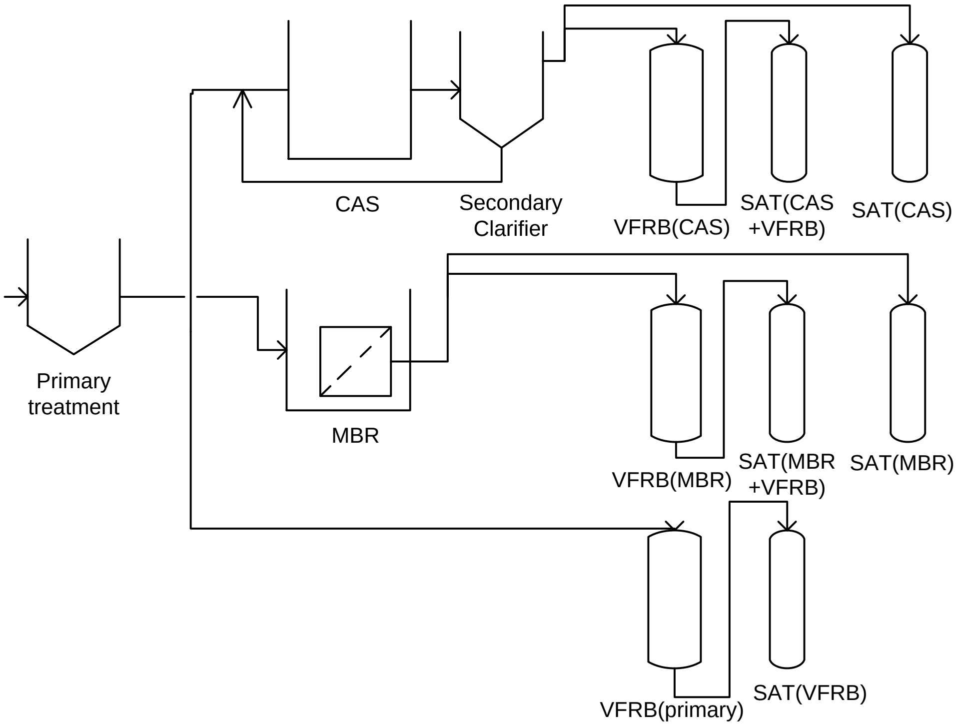

Figure 6-1 Scheme of pilot plant.

6.2.2 Metal Analysis

The samples from the primary treatment and the pilot plant were filtered and concentrated nitric acid was added at 5 % volume of matrix compounds. In order to avoid and reduce instrument contamination and interference, samples were diluted by a factor of 20. The samples were analysed using inductively coupled plasma mass spectroscopy (Perkin Elmer SCIEX ELAN 9000 ICP-MS and CETAC ASX-510 autosampler). Reproducibility was assessed by analysing four times the triplicates and variability over time was evaluated by taking primary and CAS samples every 2 d for 10 d at different hour of the day each time. The reproducibility analysis showed less than 2 % error except for CAS effluent where up to 8.7 % error was obtained. Major variations in metal concentration over time were observed in the primary effluent: from -4.0 % to +9.2 % error. However, the variability remained below 5 % for the other effluents.

6.3 Pilot plant general performance and influent metal concentration

6.3.1 General performance

Nitrification was similar in CAS and MBR, with ammonium in effluents of both processes below 0.8 $mgNH_4\text{-}N.L^{-1}$ with mean 97 % removal, and between 20 and 25 $mgNO_3\text{-}N.L^{-1}$ (Table 6-1). However COD removal in MBR was significantly higher than in CAS, respectively at 89 and 79 % at 6 d SRT. Membrane filtration was undeniably more efficient than CAS clarification for organics and solids removal. Indeed, solid content and turbidity remained below 1 $mg.L^{-1}$ and 1 NTU in MBR effluent under all operating conditions, while in CAS solid content and turbidity were respectively 18 $mg.L^{-1}$ and 9 NTU, yet achieving 88 % solids removal and 95 % turbidity removal.

A significant pH drop, down to 4.4, occurred in the MBR effluent at an SRT of 12 d, compared with a pH of 7.6 in the influent. It is suggested that unknown strongly-acidic compounds present in the influent and retained by the membrane altering the pH in the MBR and not in the CAS, where these compounds were presumably coated by the larger flocs during clarification.

The performance study also highlighted the benefits of VFRB as an additional barrier to certain pollutants (Table 6-1). VFRB provided further nitrification and particulate removal after CAS, while it degraded MBR effluent quality for solids and bacteria.

The SRT affected mainly the COD removal during the pre-treatment but did not correlate with SAT performance. However, SAT(CAS) effluent quality improved over the 18 months of column experiments , while SAT(MBR) effluent was degraded over time. Overall, SAT demonstrated high removals of solids and bacteria after only one metre of soil passage and pH was increased up to 1 unit of pH after SAT soil passage. SAT fed with CAS set up at 6 d SRT presented high autotrophic removal of TN and almost complete removal of TP. This high removal was associated with low infiltration rate and high pH.

Table 6-1 Pilot plant effluent quality (with minimum value of pH in MBR effluent in brackets).

	COD			**NH_4**			**PO_4**			**Turbidity**			**Total *Coliforms***			**pH**		
	[$mgO_2.L^{-1}$]			[$mgN.L^{-1}$]			[$mgP.L^{-1}$]			[NTU]			[$CFU.mL^{-1}$]					
SRT [d]	6	12	20	6	12	20	6	12	20	6	12	20	6	12	20	6	12	20
Primary effluent	236	508	329	28.4	27.8	27.0	4.3	11.7	7.8	120	271	184	3,E +05	2,E +05	2,E +05	7.6	7.6	7.8
MBR	26	42	32	1.2	1.0	0.7	4.0	6.7	4.8	0.7	0.9	0.5	4	0	3	6.9 [5.4]	6.1 [4.4]	6.5 [5.8]
VFRB (MBR)	28	37	21	0.7	0.3	0.8	4.2	5.0	4.5	1.9	1.6	0.9	276	105	214	6.9	7.1	6.9
SAT (MBR)	24	22	22	1.0	0.1	0.5	3.5	6.3	6.0	0.5	0.5	0.6	2	1	1	7.7	7.7	7.7
SAT (MBR +VFRB)	27	22	18	0.2	0.1	0.1	2.8	4.9	5.4	0.5	0.7	0.7	1	13	4	7.8	7.8	7.9
CAS	49	69	46	1.1	2.8	0.7	4,4	5.4	5.3	9.0	9.0	9.2	2,E +04	3,E +03	2,E +03	7.3	7.4	7.2
VFRB (CAS)	27	41	23	1.3	1.4	0.4	3.9	5.8	4.6	1.6	2.1	2.7	238	235	429	7.0	7.1	7.0
SAT (CAS)	38	34	21	0.8	0.3	0.0	0.1	4.8	6.3	0.9	0.6	1.1	43	1	1	8.3	7.8	7.7
SAT (CAS +VFRB)	19	26	20	0.3	0.3	0.0	1.8	3.9	5.1	0.6	0,8	0,5	108	4	5	7.9	7.7	7.9
VFRB (primary)	48	59	59	1.5	1.4	1.1	4.1	6.3	5.4	7.1	5,2	7,0	2,E +04	3,E +03	1,E +04	7.0	6.7	7.1
SAT (VFRB)	24	33	23	0.8	0.4	0.3	1.6	3.9	7.0	0.9	0,8	0,8	28	183	92	7.9	7.9	7.8

6.3.2 Influent metal content

The primary sedimentation tanks provided effluent quality and metals content within the range of low strength domestic wastewater despite the presence of industrial activities on campus (Oliver and Cosgrove, 1974; Patterson *et al.*, 1983) (Table 6-2). It is noticeable as well that Al and Cd were almost absent from the soluble fraction of the primary effluent, and Co and Pb concentrations were as low as 1.0 and 0.8 ppb respectively in average. As a results of some soluble metal content being very low or absent in the primary and final effluents, the metals Cd, Co and Pb will not be discussed in this study.

Table 6-2 Primary effluent quality.

SRT corresponding period [d]	Parameters [mg.L^{-1}]		Metals [ppb]								
	COD	TSS	Al	Cd	Co	Cr	Cu	Mn	Ni	Pb	Zn
6	236	81.4	0	0.0	1.2	4	25	113	36	1.0	180
12	508	141	0	0.0	1.0	6	22	99	20	1.0	260
20	329	145.8	0	0.1	0.9	11	20	65	0	0.5	380

6.4 Metal removal in SAT pre-treatment

Metal removal mechanisms in CAS and MBR include entrapment and sorption onto flocs followed by settlement or microfiltration, biodegradation, metal accumulation in cells and volatilisation (Brown and Lester, 1979). In VFRB, bioaccumulation in micro-organisms, reeds uptake, biodegradation and soil adsorption are the main pathways for metal removal (Lesage *et al.*, 2007). Goldstone *et al.*, (1990), reported different behaviour patterns between the metals during wastewater treatment in terms of removal efficiency, soluble fraction, implying solubilisation and soluble form removal, and factors influencing removal such as organics and sludge characteristics.

6.4.1 Chromium

Overall, Cr content in the effluents ranged between 2.4 and 6.3 ppb, except for VFRB(MBR) at 12 d SRT with only 0.5 ppb and at 20 d with almost 15 ppb (Figure 6-2). Chromium removal efficiency increased with influent content and SRT with the exception of CAS at 12 d SRT where no removal was achieved. Chromium removal in CAS at 20 d SRT accounted for 64 % in accord with Sterritt and Lester, (1981), who found removals above 50 %. In this case, removal was higher than expected as the influent content was only about 11 ppb. Oliver and Cosgrove, (1974), observed no more removal with influent below 10 ppb. In the MBR, only 43 % Cr was removed which was lower than in the study of Fatome *et al.*, (2005), as expected since they used ultra-filtration and longer SRT. It was however in agreement with Malamis *et al.*, (2009), who reported removal between 43.2 and 69.2 % of an influent with 320 ppm. The lower Cr removal in MBR at 20 d SRT compared to CAS, respectively 43 and 64 %, can be explained by the higher oxygen transfer in MBR due to the smaller floc size, with 39 compared to 252 µm for CAS. Oxygen is known to interfere with the biological reduction of Cr(VI) (Komori *et al.*, 1990; Chen and Gu, 2005). However, Sterritt *et al.*, (1981), demonstrated little removal of Cr(IV) by precipitation. Hence it is predicted that the larger flocs in CAS would promote removal by adsorption onto flocs and sedimentation compared to the MBR. Other studies have highlighted the mechanism of chemisorption of Cr to sulphide and carbonate compounds and the role of bacterial extracellular polymers in metal removal (Dugan and Pickman, 1972; Rivera, 1983).

For CAS, removal increased as floc size decreased and temperature increased. To illustrate, removal of 64 % was associated with a floc size of 252 µm and a temperature of 13.2 °C. Conversely at a lower temperature of 5.4 °C the floc size was 400 µm and removal -3 %. This suggested the predominance of Cr binding with organics and biodegradation over other mechanisms, in agreement with Chen and Gu, (2005).

For MBR, increased removal was associated with increased COD and particularly floc size. For example, removal of 65 % was achieved with an influent COD content of 508 mg.L^{-1} and a floc size of 224 µm.

VFRB(primary) supplied the best removal for Cr compared to the other reed beds, between 36 and 79 %, yet below the 99 % removal reported by Tiglyene *et al.*, (2005), however considering total chromium removal. This was probably related to the higher influent content for VFRB(primary) of 4 to 11 ppb compared with 2 to 6 ppb for the other reed bed configurations. VFRB(CAS) and VFRB(MBR) had positive removals only at 12 d SRT with 29 and 74 %. Cr was negatively correlated with pH in VFRBs.

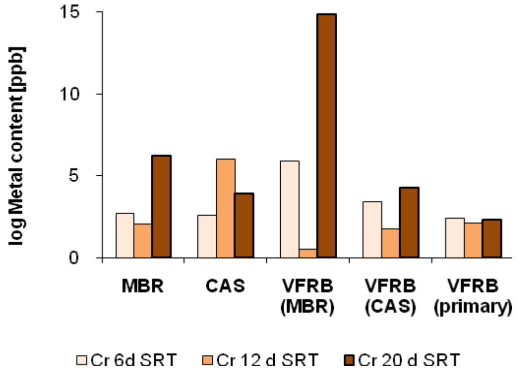

Figure 6-2 Cr content in pre-treatment effluents.

6.4.2 Copper

Cu content in CAS and MBR effluents remained below 10 ppb except at 12 d SRT with 20 and 12 ppb respectively (Figure 6-3). At 6 and 20 d SRT both CAS and MBR provided better removal than in most of the literature. Copper removal in CAS at 12 d SRT was in the range of the literature below 40 %, while in MBR it was below the range specific to microfiltration of Innocenti *et al.*, (2002), and Fatone *et al.*, (2005), with only 46 % compared to more than 70 %. Copper removal in CAS showed the same pattern as Cr removal with more pronounced removal at 20 d SRT. This may be associated with the higher temperature of

12.2 °C and the smaller floc size with 252 µm at 20 d SRT promoting biological removal rather than floc adsorption (Table 6-1). The best Cu removal was observed at 6 d SRT with 90 %, when pH of 6.9 and the ORP more favourable to oxidation process at 200 mV.

Copper content in the VFRB effluents was increased relative to the feed content and correlated negatively with nitrate content in the effluent. In VFRB(MBR), a peak of 29.6 ppb was observed at 20 d SRT compared to 11.9 and 10.1ppb at 6 and 12 d SRT respectively. The Cu content in VFRB(CAS) effluent was lower than in the VFRB(MBR) except at 6 d SRT respectively with 22.4, 5.9 and 4.8 ppb at 6, 12 and 20 d SRT. The VFRB(primary) effluent had higher copper content at above 30 ppb than the tertiary VFRBs, where average values were 11.0 and 17.2 ppb respectively for VFRB(CAS) and VFRB(MBR).

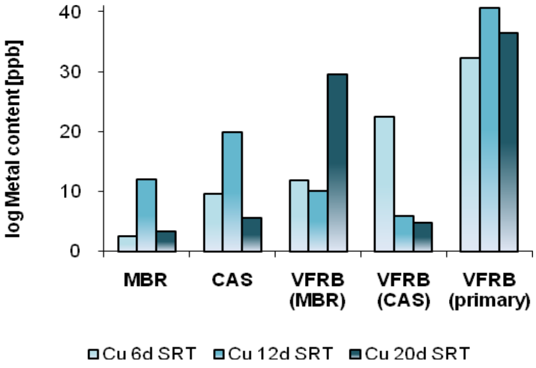

Figure 6-3 Cu content in pre-treatment effluents.

6.4.3 Manganese

Mn content was the lowest in VFRB(CAS) below 30 ppb and below 50 ppb in the other treatments except MBR at 12 d SRT with 81 ppb, VFRB(MBR) at 20 d SRT with 354 ppb and VFRB(primary) with 210, 125 and 66 ppb respectively after 6, 12 and 18 months (Figure 6-4). Mn removal in CAS ranged from 31 to 58 % which is in accord with Stephenson and Lester, (1987), but higher than Sterritt and Lester, (1981), and Kempton *et al.*, (1983). The MBR provided a little bit more removal at 6 and 20 d SRT with 65 and 51 % than CAS at 61 and

58 %. At 12 d SRT, only 18 % Mn was removed in the MBR and it was related to the pH drop in the MBR.

VFRB(MBR) provided Mn removal between 22 and 44 % for 6 and 12 d SRT in accord with Obarska-Pempkowiak and Klimkowska, (1999). At 20 d SRT, Mn increased, with an effluent content of 354 ppb. Manganese content remained below 26.4 ppb in VFRB(CAS) and encountered less variation than VFRB(MBR) or VFRB(primary). It provided the best removal with 33, 68 and 43 % for 6, 12 and 20 d SRT respectively. VFRB(primary) only removed Mn during the period corresponding to 12 d SRT with 33 % removal. For the other periods corresponding to 6 and 20 d SRT, Mn increased to 210 and 125 ppb respectively in the VFRB(primary) effluent.

Manganese removal in CAS and MBR was correlated to the Mn feed content. Higher Mn loading with 113 ppb was present at 6 d SRT with a higher Mn removal in both CAS and MBR respectively at 58 and 65 %, compared to the other SRTs, when removals varied from 18 to 51 %.

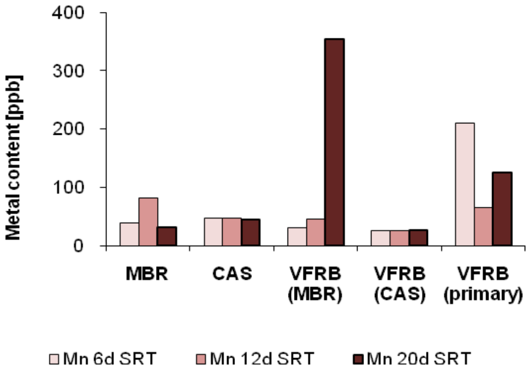

Figure 6-4 Mn content in pre-treatment effluents.

6.4.4 Nickel

Overall, Ni content remained below 35 ppb and was even absent in VFRB(CAS) except in CAS at 6 d SRT with a peak of 77.4 ppb (Figure 6-5). Nickel removal showed large variations in efficiency in both CAS and MBR. Ni removal of 100 % was achieved at 12 d SRT for both processes, while at 6 d SRT removals were –115 % and 8 % for CAS and MBR respectively. At 20 d SRT, no nickel was present in the influent and only CAS and VFRB(MBR) released Ni with 26.8 and 13.0 ppb. This can be explained by microbial processes releasing nickel into the matrix or by solubilisation of the metal during secondary treatment (Lester, 1987; Nielsen and Hrudey, 1983). Given the extent of the variations, no correlation could be made with the analysed parameters.

Nickel was absent from VFRB(CAS) effluent at all SRT, with concentrations only of 1.7 ppb in VFRB(primary) effluent, which corresponded to 95 % removal. However, in VFRB(MBR) effluent, Ni content was comparable to MBR effluent content with 32.0 ppb at 6 d SRT and 13.0 ppb at 20 d SRT when it was absent from the MBR effluent. Nickel content and removal were in accord with literature, but the same correlation with temperature and pH could not be demonstrated (Sasaki *et al.*, 2003; Norrstrom and Jacks, 1998; Sanford, 1999).

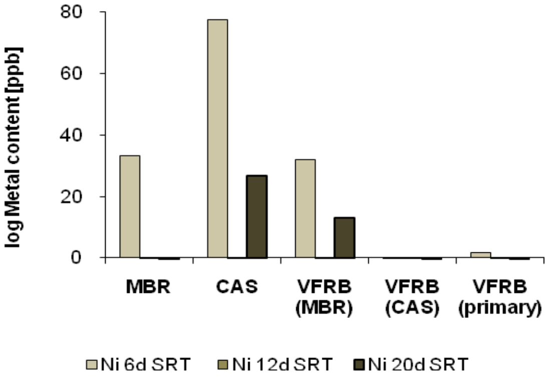

Figure 6-5 Ni content in pre-treatment effluents.

6.4.5 Zinc

Zn content showed a similar pattern between the treatments influenced by SRT changes with a range of 100 to 200 ppb at 6 d, 450 to 600 at 12 d and 300 to 450 at 20 d, while in VFRB(primary), Zn content diminished over time with 1350, 1056 and 902 after 6, 12 and 18 months (Figure 6-6). Zinc removal in CAS and MBR ranged between 0 and 39 % with no removal at 12 d SRT and highest removal at 6 d SRT. The removal range and the reduction in performance at 12 d SRT tally with the results from Kempton *et al.*, (1983). At 6 d SRT, the removal was higher than at 20 d SRT as previously found by various authors (Kempton *et al.,* 1983; Stoveland and Lester 1980; Sterritt and Lester, 1981). The MBR removal was lower than reported by with Fatone *et al.*, (2005), who found more than 90 % removal, though using a membrane of pore size 0.04 µm, compared with the micro-filtration of 0.08 µm in this study.

This pattern was associated with higher floc size and low temperature of 5.4 °C for MBR and CAS at 12 d SRT. Low temperatures are known to adversely affect zinc removal by changing microbial characteristics and consequently adsorption onto flocs (Dugan and Pickman, 1972; Rivera, 1983). Nielsen and Hrudey, (1983), also observed extensive variation of performance, from 4 to 75 % removal, with effluent content between 120 and 580 ppb. Effluent concentrations in this study were in this range at 330 and 310 ppb for CAS and MBR respectively.

Zinc leaked from all VFRB. VFRB(CAS) and VFRB(MBR) had lower Zn content at 6 d SRT with less than 200 ppb compared to 12 and 20 d SRT when Zn content was above 400 ppb. In VFRB(primary) the effluent content was initially up to 10 times the primary effluent content, then lowered with time but never fell below 900 ppb. Zn in VFRBs effluents was correlated with turbidity and total *Coliforms.*

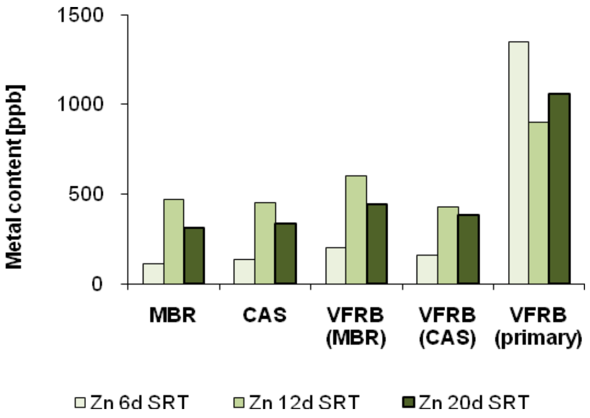

Figure 6-6 Zn content in pre-treatment effluents.

6.5 Metal removal in SAT

6.5.1 Chromium

For SAT influent, Cr concentration ranged from 0.5 to 14.8 ppb respectively for VFRB(MBR) at 12 and 20 d SRT. Meanwhile, maximum and minimum levels in the effluents were 0.3 ppb for SAT(VFRB) after 18 months and 6.4 ppb for SAT(CAS+VFRB) at 6 d SRT (Figure 6-7).

At 6 d SRT, only SAT(MBR+VFRB) offered Cr removal with 40 %, but all SAT effluents remained below 4 ppb. At 12 d SRT, SAT(MBR) and SAT(CAS) were the only SAT removing Cr, by 21 and 85 % respectively, producing effluents below 1 ppb, compared to the other SATs where Cr effluent content was between 2.3 and 3.5 ppb. However, at 20 d SRT only SAT(CAS) had negative removal, yet with a low effluent content of 4.20 ppb. SAT(MBR) removal was also low at 20 % compared to more than 73 % in the other SATs. The best effluents qualities were provided by SAT(CAS+VFRB) and SAT(VFRB) with concentrations of 0.50 and 0.29 ppb. Cr content was suspected to be correlated with total *Coliforms* and turbidity in SAT effluent but more data should be obtained to confirm this hypothesis. Differences in removal between this study and that of Aziz *et al.,* (2008), may be attributed to corresponding variation in influent concentration at 6.8 ppb and 2 000 ppb respectively. At the latter level, metal precipitation in addition to sorption onto carbonate is likely (Xu *et al.,* 1996; Cheng *et al.*, 1998). According to Hwang *et al.*, (2002), and Oh *et al.*, (2007), reactions between Cr and Fe should be enhanced in the presence of silica and sand, with resolubilisation expected to be difficult. However, despite this study operating under such conditions, Cr leakage was observed. The negative correlation of Cr removal with Mn suggests competition with this metal as it adsorbs onto ferric oxide partially irreversibly (Stollenwerk, 1994).

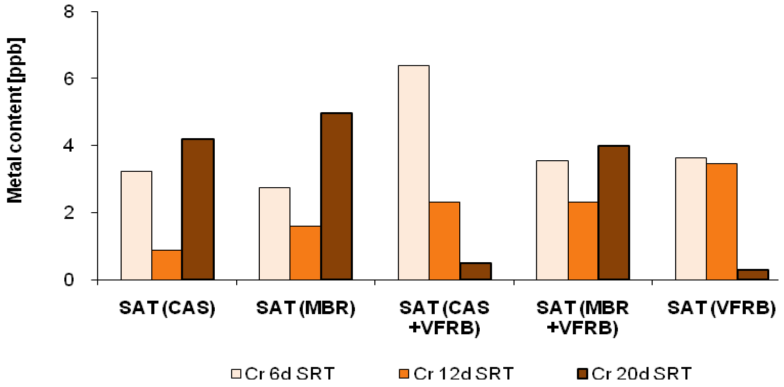

Figure 6-7 Cr content in SAT effluents.

6.5.2 Copper

SAT(CAS) at 20 d SRT provided the lowest Cu effluent content at 3.70 ppb while SAT(VFRB) provided the highest concentration of 21.6 ppb (Figure 6-8). At 6 d SRT, negative removals were observed in SAT(MBR) and SAT(MBR+VFRB), 25 % was removed in SAT(CAS), while removal in the other SRT was about 55 %. At 12 d SRT, Cu leakage occurred in SAT(CAS+VFRB), low removal was obtained by SAT(MBR+VFRB) and SAT(CAS) at 13 and 8 % respectively, while higher removals of 56 and 76 % were observed in SAT(MBR) and SAT(VFRB) respectively. At 20 d SRT, removal ranged between 34 and 66 % with the exception of SAT(MBR) and SAT(CAS+VFRB) where slight increases were recorded.

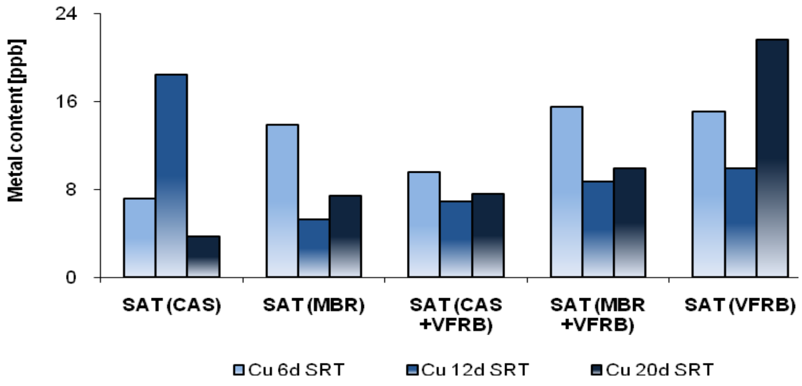

Figure 6-8 Cu content in SAT effluents.

6.5.3 Manganese

SAT(MBR) provided the lowest Mn effluent content, of 15.2 ppb at 20 d SRT, though also the lowest removal with 0.5 % at 6 d SRT (Figure 6-9). SAT(MBR+VFRB) had the highest removal of 91 % at 20 d SRT. Removal was negative for SAT(MBR+VFRB) and SAT(CAS+VFRB) at 6 d SRT and SAT(CAS+VFRB) at 20 d SRT. At 6 d SRT, SAT(VFRB) and SAT(MBR+VFRB) had the highest effluent content with 91 and 84 ppb, while for SAT(CAS) the level was below 20 ppb and around 40 ppb for the other SATs. Mn content ranged between 16.5 and 33.6 ppb at 12 d SRT, and between 15.2 and 46 ppb at 20 d SRT. Mn content at 20 d SRT was correlated to the pH and negatively to phosphate content in SAT effluents.

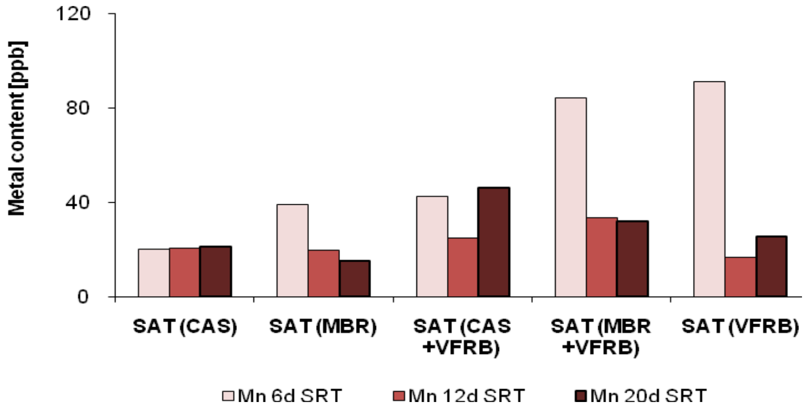

Figure 6-9 Mn content in SAT effluents.

6.5.4 Zinc

Zn content remained between 30 and 73 ppb at 6 d SRT in all treatments while at 12 d SAT(CAS) and SAT(VFRB) reached peaks above 135 ppb and Zn was absent in SAT(CAS+VFRB) and SAT(MBR). At 20 d SRT, Zn content was still higher in SAT(CAS) with 117 ppb, remained around 80 ppb in SAT(MBR+VFRB) and SAT(VFRB) and was lower or even absent in SAT(MBR) and SAT(CAS+VFRB) (Figure 6-10).

SAT provided good removal of Zn with values between 60 and 100 % in all samples. SAT(CAS+VFRB) removed all Zn at all SRTs. However, high concentration of 135.8 and 165.4 ppb were observed in SAT(CAS) and SAT(VFRB) effluent at 12 d SRT. The high removal of Zn is comparable to data quoted by Aziz *et al.*, (2008), who give these metals' removal by contact with limestone of between 85 and 98 %. Vanbroekhoven *et al.*, (2007), referred to the effect of increased pH on Zn removal though they considered that the increase in pH was caused by the conjunction of nitrate reduction conditions and presence of organic matter. In our study, only SAT(CAS) at 6 d SRT fulfilled these conditions, though all SATs offered similarly high removal. However, every SATs provided buffering and an increase in pH after soil passage. Hence the presence of any relationship between pH and Zn removal is probably masked. Zn content at 12 d SRT was related to the pH in SAT effluent. It is worth noting that at a pH of 7.5 it is considered Zn adsorption should not be limited (Coston *et al.*, 1995).

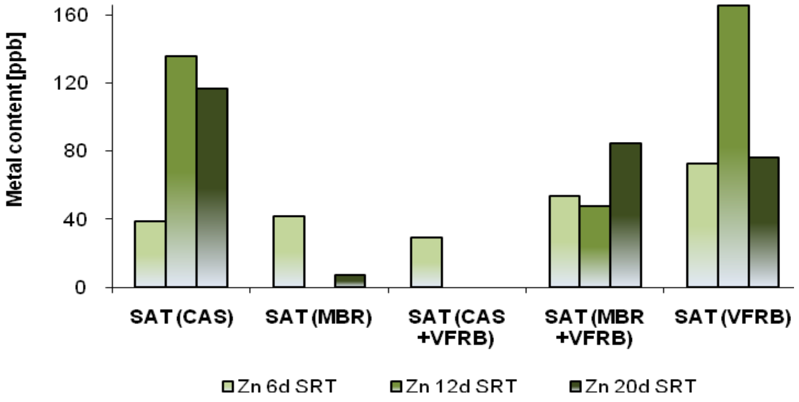

Figure 6-10 Zn content in SAT effluents.

6.5.5 Aluminium and Nickel

Aluminium and Nickel were only found in SAT(MBR) effluent with Al contents of 13.2 and 121.8 ppb at 6 and 12 d SRT and Ni content of 51.4 ppb at 6 d SRT (Figure 6-11). For the other SATs, when Ni was present in the SAT feed, removals were always of 100 %, in accord with Aziz *et al.*, (2008), who reported removal by contact with limestone as between 85 and 98 %. This also tallies with Christensen *et al.*, (1996), who found that Ni mobility in a sandy aquifer was significant only at pH below 5. This was verified for Ni as well, with complete removal with the exception of SAT(MBR), when the MBR effluent pH was below 6 compared with a mean of 7.1 in the other processes. Aluminium release from SAT(MBR) at 6 and 12 d SRT was atypical as Al was not detected anywhere else in the flow sheet. This implied the dissolution of Al already present in SAT soil, probably in the form of $Al(OH)_3$ given the pH drop from an average of 6.5 to a minimum of 4.3 in MBR effluent (Stollenwerk *et al.*, 1994).

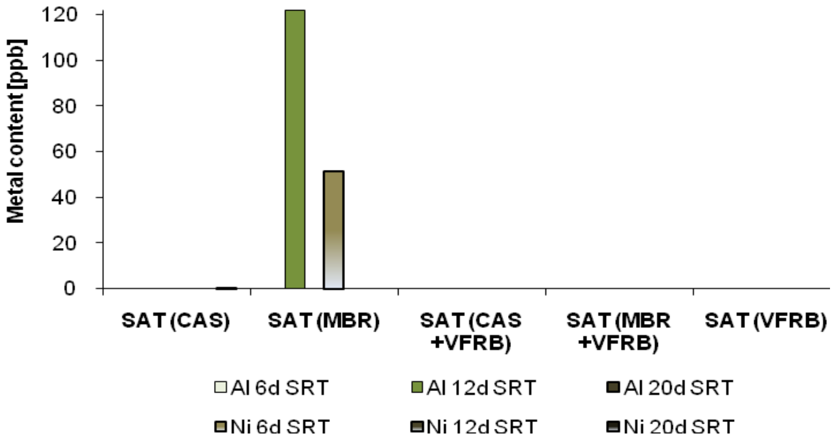

Figure 6-11 Al and Ni content in SAT effluents.

6.6 Implication of metal effluent quality and removal for reuse

Considering the average removals and effluent contents of total metal, the treatment train VFRB(CAS) provided the best option of pre-treatment followed by MBR and CAS respectively with 13, 11 and 2 % removal. Nevertheless, removal of total metals after SAT ranged between 58 and 88 % respectively for SAT(VFRB) and SAT(CAS+VFRB).

Both CAS and MBR complied towards drinking water and irrigation reuse standards for all metals, with the exception of nickel at 6 d SRT and in CAS effluent at 20 d SRT for drinking water reuse. However, primary effluent was already complying for the same metals, namely Zn, Cu and Cr. CAS removed half the metal relative to the MBR but the same pattern was detected with a better removal of 24 % at 6 d compared to 13 % at 20 d and with a 31 % increase at 12 d SRT. VFRB(primary) did not improve or even reduced the compliance of primary effluent towards water standards. Indeed, VFRB(primary) failed to comply with irrigation reuse standards for the first 6 months, producing 210 ppb of Mn, just above the limit of 200 ppb. However, this treatment did comply with the Ni limit of 20 ppb with a maximum of 1.7 ppb, whereas primary effluent did not at 36 and 20 ppb after 6 and 18 months. VFRB(primary) presented increased level of total metals for the three sampling times leaving the effluent with a very high soluble metal content between 1015 and 1601 ppb.

Overall, the MBR provided better removals of all metals than the other treatments. Nevertheless, as problems occurred in the MBR leading to significant drop in pH at 12 d SRT, the CAS, more stable with the changes in SRT, provided better effluent quality for metals. No direct relationship could be made between SRT and metal removal, which correlated better with external conditions such as influent concentration, temperature and pH. Nonetheless, SRT is involved in floc size variation, which was linked to metal removal in this study, and SRT is also known to affect the type and concentration of organics, involved in metal binding, and metal solubility (Rossin *et al.*, 1982; Oliver and Cosgrove, 1974).

This study focused on the soluble fraction of metals and even where particulate metal compounds have been removed or solubilised after primary treatment, new complexes of metal with organics, flocs or precipitates could still leak from the CAS clarifier (Barth *et al.*, 1965; Chen *et al.*, 1974). Another concern is to assess the capacity of the biomass to accumulate metals and hence subsequently release them, either sporadically or regularly. When considering the maximum release percentage as well as the absolute value, the MBR set up at 12 d SRT was able to liberate up to 700 µgZn.gMLSS^{-1}.

CAS and MBR represent similar risk of groundwater contamination as metal transport is promoted by dissolved macromolecules and colloids such as bacterial polymers that bind the metal to the aqueous phase (Chudoba *et al.,* 1986; McCarthy *et al.,* 1993; Chen *et al.*, 1995). For this reason, the addition of soil based tertiary treatment was examined to counter the effect of influent variability and biological release of accumulated metal from CAS and MBR.

VFRB(CAS) complied completely with both sets of standards, whereas CAS alone had failed both standards at 6 d SRT. VFRB(CAS) offered better removal as a unit than the other reed beds tested, with 13 and 22 % removals for 12 and 6 d SRT, and -1 % at 20 d SRT. Heightened levels of total metal was a trend of VFRB(MBR) with 52, 17 and 138 % increases respectively for 6, 12 and 20 d SRT. VFRB(MBR) failed to comply with drinking water standards for Ni and with irrigation standards for Mn, producing levels of 32 ppb at 6 d SRT and 354 ppb at 20 d SRT respectively.

In terms of SAT, all treatment trains complied except SAT(MBR) at 6 d SRT. Indeed, SAT(MBR) at 6d SRT did not comply with drinking water standards with 122 ppb of Al compared to the limit of 100 ppb. It also did not pass the drinking water standard for Ni with 51.4 ppb at 6d SRT. Nevertheless, SAT compensated for the metal increase in MBR at 12 d SRT with 74 % removal of total metal. Highest removal at 90 % in SAT(MBR) was also obtained at 6 d SRT, with the lowest removal of 13% at 20 d SRT. The same trend was observed in SAT(CAS), providing a constant removal of total metal between 65 and 74 % for 20 and 6 d SRT.

Overall VFRB followed by SAT provided between 48 and 74 % removal, corresponding to 81 and 90 % removal in the SAT unit. SAT(VFRB(MBR)) removed between 45 and 86 % of total metal at 6 and 12 d SRT. SAT(VFRB) after 12 months was even more promising, with a total metal removal between 58 and 96 %. It was hence the best treatment train for total metals removal, up to 97 % for the same period.

Lin *et al.*, (2004), addressed some concerns about the long term decline of metal removal by this type of SAT soil, caused by the decomposition of applied organic matter leading to carbonate and manganese oxide dissolution and hence diminishing the soils affinity for metal. In this study only SAT(CAS) showed increased Zn and Cu effluent content with time, while for the other treatments the trend over 18 months was even a decrease in metal effluent content. This presumably indicates the discussed dissolution pathways were not significant here.

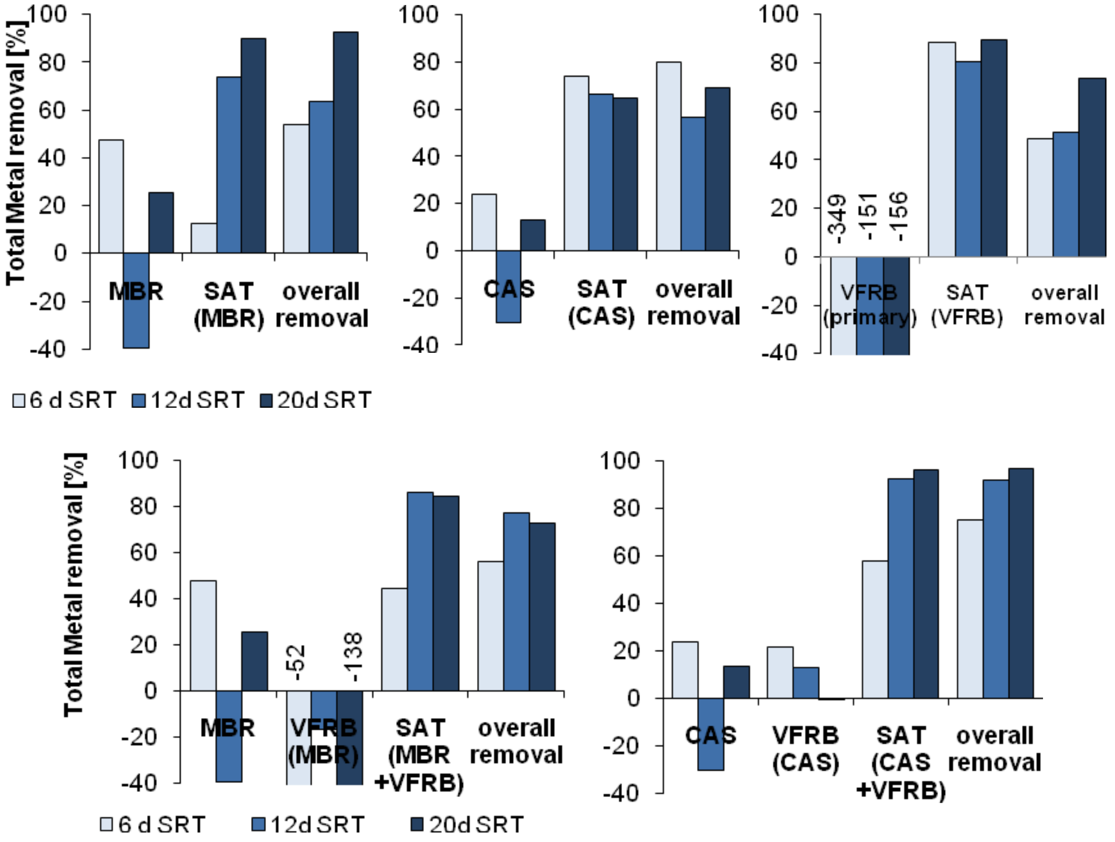

Figure 6-12 Percentage removal of total metal for each treatment unit and all flow sheets, at 6, 12 and 20d SRT or corresponding period of time for VFRB(primary)and SAT(VFRB) (with data label for removals below -40%).

6.7 Conclusions

The effluents from all the pre-treatments complied with irrigation reuse standards as well as drinking water standards with the exception of nickel.

Overall the MBR provided higher metals removals than CAS. VFRB showed metal leakage, mainly for Cu and Zn where content increased consistently in all reed beds at all SRTs. Overall, SAT improved all effluents despite metal leakage in VFRB, and especially for VFRB(CAS) effluent, with between 87 and 93 % metal removal and total metal effluent content below 90 ppb. Only SAT(MBR) raised concerns with Ni and Al leakages.

This study illustrated the possible effect of COD, pH, ORP and floc size variations on metal removal, while no direct correlation with SRT was observed. Indeed, the SRT and time were not correlated with metal removal in VFRB but other correlations were observed for Cr with pH, Cu with nitrate and turbidity and Zn with turbidity and total *Coliforms*. SAT did not show any correlation with SRT or time either. However, Zn was correlated with pH, Mn with pH and phosphate and Cr with total *Coliforms* and turbidity.

Further research is recommended for all metals on comparing CAS and MBR and including metal uptake and release from particulate biomass as well as MBR operated with a microfiltration membrane. Also, the potential for bioaccumulation of metals and the associated risks of failure should be determined.

Only SAT(CAS) showed Zn and Cu effluent content increase with time, while for the rest the trend over 18 months was even a decrease in metal effluent content. However, a trend over the experiment may not be extrapolated to decades of full scale service of SAT so longer experiments or sorption kinetics tests are recommended.

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7 OVERALL DISCUSSION

The general aim of this work was to establish the potential role of VFRB as an upgrade technology option in comparison to using MBRs. Overall, whilst differences in the water qualities remained, the work outlined within this thesis demonstrates that the use of VFRB can be considered a suitable alternative to upgrading treatment works to MBRs. Specific benefits when using a VFRB were identified in terms of improved removal, reduced SAT clogging and perhaps more importantly a greater robustness which comes about from linking technologies in a treatment train (Table 7-1).

Table 7-1 Comparison of tertiary VFRB versus MBR as SAT pre-treatments.

Chapters	***Focus***	***Pros***	***Cons***
Chapt. 2	Literature	Reduced Capex and Opex Additional barrier	Risk of nitrate pollution Risk of reed bed soil clogging
Chapt. 3	SAT pre-treatment	Better overall removals Buffers influent variations Provides better N removal and pH and conductivity control	MBR remained the best treatment for bacteria and solids removals. At short SRT MBR perform better than tertiary VFRB
Chapt. 4	Impact on SAT	Reduced clogging of SAT soil Buffers temperature impact	Lower infiltration rates
Chapt. 5	Phosphorus	Better P removal	Reduced P capacity in SAT than MBR and CAS effluents
Chapt. 6	Metals	Complied for drinking water standards for metal after SAT	Metal release after VFRB

Specifically, the use of tertiary VFRB was shown to enhance reclaim water quality, especially after CAS, and to reduce SAT clogging compared to direct application of MBR effluent onto the SAT (Table 7-1). This confirms literature suggestions concerning the potential benefit of the technology especially where the removal of pollutants, which are more difficult to attenuate in SAT and that can cause soil clogging is critical (Chapter 2). Specific to the conditions of the trials outlined in this study, additional benefits of VFRB after CAS were seen in terms of nitrification and particulate removal (Chapter 3) with VFRB(CAS) effluent quality being similar to or even better than that from the MBR for few parameters. Importantly, VFRB was also shown to buffer upstream variations (Table 7-2), which is potentially of great importance to managed aquifer recharge schemes and matches to one of the key benefits of using membrane based processes for reuse applications (Judd and Jefferson, 2003).

Weighted against such benefits were lower infiltration rates, maintenance issues to do with the VFRB and a poorer performance in terms of solids and bacteria removal. However, this is to be expected as the membrane in MBRs acts as an absolute barrier which cannot be easily replicated through porous filtration systems. Consequently, comparison of the two technologies at a treatment pathway level indicates that the additional solids removal and biological aerobic activity provided by the VFRB can offset much of the benefits of intensification generated by MBRs. However, the microbial communities and associated activity levels must be different due to differences in microbial selection pressures and loading rates. This is manifest in the observation that the MBR effluent contained soluble pollutants that could not be filtered by either a VFRB or SAT due in part to the short retention time generated by their mobility. Additionally, differences in solids and organic profiles from the two options generated different metal removal profiles with periodic leaching of Cu and Zn (Chapter 6) from the VFRB that was not observed in the case of the MBR. However, the opposite feature was observed when including SAT with better metal removal in VFRB trains than MBR train.

Overall this indicates a balance between considering the SAT as a treatment process and the infiltration rates possible. The principle consequence of this is the land requirement when designing the initial system which would need to be considered within a total cost and energy appraisal. Whilst cost and energy assessments were outside the scope of the current investigation, information available in the literature would suggest the use of MBR to be limited by capex and opex where typical energy demands for operating MBRs plants can exceed 1 KWh.m^{-3} (Judd, 2006). Comparatively then, VFRB offer a low cost, low energy upgrade option which does not even need to be balanced against available land on the contrary to horizontal reed beds. Given that the use of VFRB enhances infiltration rates compared to CAS or reduces long term clogging compared to a membrane, a balance is possible in terms of total SAT land required that would need further investigation to validate.

Table 7-2 Suitability of the different treatment trains for SAT and irrigation with severe restrictions.

Pre-Treatment	Compliance SAT [1]/[2]-irrigation [%]	Clogging prevention	Robustness	Reliability	Complexity	Cost
MBR	**81/91 – 79 (*)**	**-**	**+++/+(**)**	**++**	**+++**	**+++**
CAS	82/87 – 78	++	+ /+++(**)	+	++	++
MBR +VFRB	**88/93 – 89**	**+**	**++**	**+++**	**+++**	**++++**
CAS +VFRB	**82/87 – 85**	**+**	**++**	**+++**	**++**	**+++**
VFRB	75/87 - 75	+	+	+	+	+

[1] prior SAT

[2] after 1 m SAT

(*) It is noticeable that the MBR was the best treatment for SAT compliance at 6 d SRT with 95 %

(**) Robustness of SAT(MBR) was high compared to the other treatments except at 6 d SRT where it was the least robust, and the opposite trend applied for SAT(CAS).

Considering the breadth of treatment trains investigated the main observed impact of technology selection was seen in relation to the infiltration rate of the

SAT which is directly related to the solids loading. To illustrate, infiltration rates of 2.6 m.d^{-1} SAT(CAS), 16.7 m.d^{-1} SAT(VFRB), 22.7 m.d^{-1} SAT(tertiary VFRB) and 46.6 m.d^{-1} SAT(MBR) were consistent with TSS levels of 24.5 mg.L^{-1} CAS effluent, 16.3 mg.L^{-1} VFRB(primary), 5.4 mg.L^{-1} tertiary VFRB and 1.0 mg.L^{-1} MBR. Whilst indicating a relatively clear relationship between infiltration rate and TSS, analysis of the variations in infiltration throughout the trials revealed an importance of temperature in this understanding. In particular, this is most evident when loading the SAT with high TSS levels such as in the SAT(CAS) and SAT(VFRB) where greater variation was observed. In cases of lower TSS loading, temperature appears less critical but a long term reduction in infiltration rate was observed. As such selection of either MBR or tertiary VFRB provides a more robust operation of the plant and reduces the need to tailor SAT operation to climatic changes.

However, this study also demonstrated that solids loading, and hence pre-treatment, did not influence long term clogging. In fact, effluents rich in bacteria, nutrients and slowly biodegradable particles, such as those from CAS and secondary VFRB, provided the ingredients for further degradation in SAT as well as longer residence times than the other effluents and thus maximised the use of the SAT as a treatment stage. This tallied with sorption isotherms experiments as the same ranking in P removal efficiencies was observed when shaking SAT soil with the different effluents for the same duration. The studied soil showed a high potential for phosphorus removal at low infiltration rate.

If little difference in compliance towards reuse standards can be observed between VFRB(MBR) and VFRB(CAS), the treatment choice for irrigation reuse is in favour of VFRB(MBR). Indeed, VFRB(MBR) effluent was shown to have an additional value when feeding crops such as lettuces in terms of growth yield compared to VFRB(CAS) effluent (Appendix V Lettuces Trial).

7.1 References

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8 CONCLUSIONS

The main conclusions from the current study are:

1. The major impact of pre-treatment selection ahead of SAT is related to the importance of TSS content on SAT removal mechanisms and infiltration rate with a general relationship between TSS loading and infiltration rate.

2. SAT fed with high TSS content effluent was especially susceptible to temperature variations requiring changes to SAT management practices.

3. Variation of SRT only indirectly affected SAT removal at low SRT where the N compounds balance was favourable to an autotrophic N removal in the case of the CAS flow sheet.

4. High P removal can be achieved with the studied soil, especially at low infiltration rate. VFRB trains did not provide the best performances for P. However the impact of P removal on soil was buffered.

5. While soluble metal was leaking from VFRBs, SAT insured compliance in all trains.

6. Hence, tertiary VFRB provides an effective alternative to the use of membrane technologies for SAT pre-treatment.

Future research

In this study tertiary VFRBs proved to be the best option for SAT and irrigation reuse. However, compliance towards reuse standards was not achieved for all parameters. Suggested future research should focus on methods to enhance removal and so investigate the potential to engineer compliance within constructed wetlands. From the experience gained throughout the project three upgrading approaches appear worth future investigation as they would resolve some of the outstanding issues observed during the current study:

1. Combination of reed beds units, such as reed beds in series or hybrid systems would enhance the overall removal of pollutants and provide an effluent quality more constant with additional buffering barriers.

2. Novel media have become available during the course on the project (Frazer-Williams, 2007) which would potentially greatly enhance the performance of the reed bed, especially towards P and metals. Work should be undertaken to ascertain their suitability and economic viability.

3. Incorporation of anoxic and anaerobic zones to provide complete nutrient removal ahead of the SAT. Whilst this is common place in many developed countries, use of such amendments is scarce in other parts of the world. Low cost methods of incorporating such treatment pathways within tertiary treatment offer great potential. One method to consider is the use of hybrid system, horizontal and vertical flow systems in series, as it will utilise the combination of treatment pathways available in each system.

Further work on SAT is also needed to establish the exact interactions between TSS content in pre-treatment effluent, temperature and SAT removal mechanisms. This could be achieved using controlled temperature trials and constant primary effluent quality. By reducing the water head of the SAT basin, the infiltration rate in SAT fed with low TSS pre-treatments, such as VFRB and MBR trains, will be reduced and is predicted to lead to enhanced and controlled P removal in the upper zone of SAT soil.

Reference

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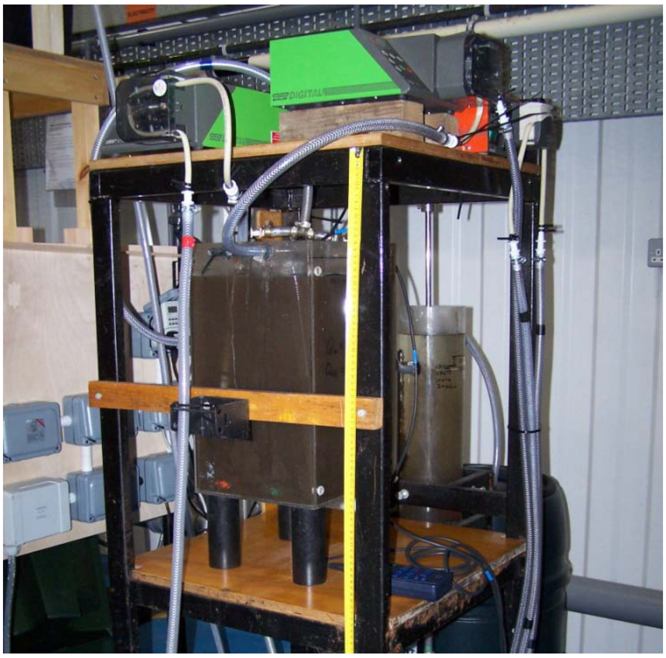
APPENDIX I Pilot Plant Pictures

MBR (left) and Sewage feed tank (right)



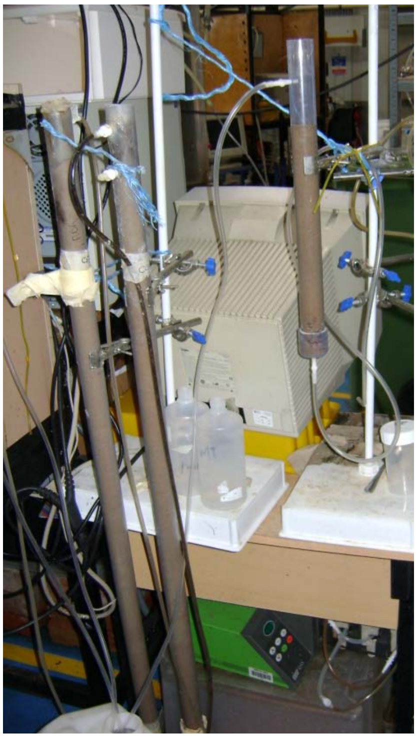
CAS

VFRB(primary)

VFRB(CAS)

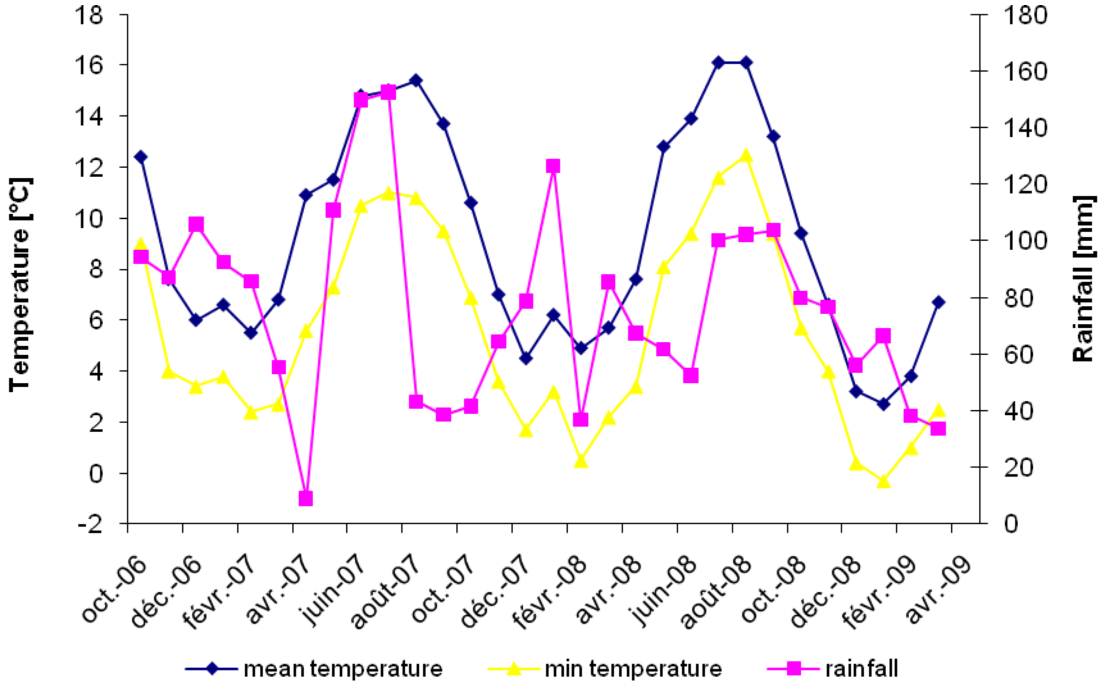
VFRB(MBR)

Harvested VFRBs



SAT columns (left)
BV column (right)

APPENDIX II Pilot Plant Data



Weather conditions

Average sulphate content and total count

SO_4 [mgS.L^{-1}]				**Total Count [CFU.mL^{-1}]**		
SRT	6	12	20	6	12	20
MBR	61	75	61	991	7	20
VFRB(MBR)	59	74	61	479	4548	3422
SAT(MBR)	61	64	98	1426	23	8
SAT(MBR+VFRB)	61	65	70	964	187	300
CAS	59	65	40	87701	10611	7358
VFRB(CAS)	56	65	52	813	7430	951
SAT(CAS)	58	68	64	155	7	119
SAT(CAS+VFRB)	59	45	70	1409	53	50
VFRB(primary)	60	92	33	33929	9718	23474
SAT(VFRB)	60	79	69	611	300	48
Primary effluent	62	91	77	1260944	1097000	825000

APPENDIX III SAT Soil Data

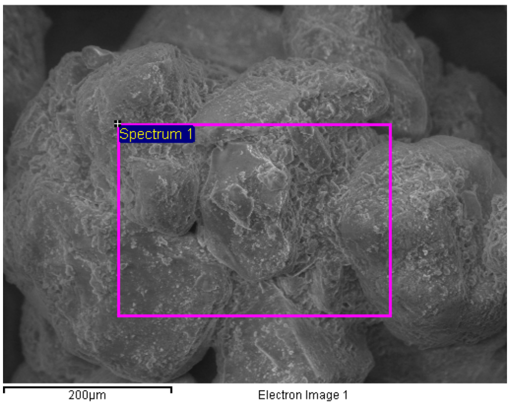
ESEM spectrum for SAT(CAS) at 20 d SRT

Spectrum processing :
No peaks omitted

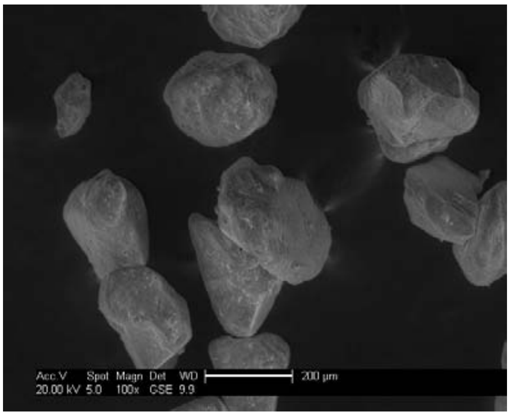
Processing option : All elements analyzed (Normalised)
Number of iterations = 5

Standard :
C CaCO3 1-Jun-1999 12:00 AM
O SiO2 23-Mar-2009 03:42 PM
Na Albite 1-Jun-1999 12:00 AM
Mg MgO 1-Jun-1999 12:00 AM
Al Al2O3 1-Jun-1999 12:00 AM
Si SiO2 23-Mar-2009 03:42 PM
P GaP 1-Jun-1999 12:00 AM
K MAD-10 Feldspar 1-Jun-1999 12:00 AM
Ca Wollastonite 1-Jun-1999 12:00 AM
Ti Ti 1-Jun-1999 12:00 AM
Fe Fe 1-Jun-1999 12:00 AM

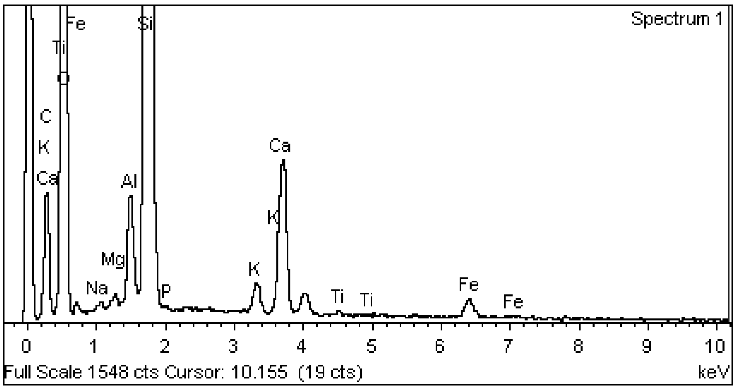
Element	*Weight%*	*Atomic%*
C K	*20.43*	*29.29*
O K	*50.32*	*54.16*
Na K	*0.23*	*0.17*
Mg K	*0.22*	*0.15*
Al K	*1.95*	*1.25*
Si K	*19.57*	*12.00*
P K	*0.10*	*0.06*
K K	*0.82*	*0.36*
Ca K	*4.83*	*2.07*
Ti K	*0.15*	*0.05*
Fe K	*1.38*	*0.43*
Totals	*100.00*	



SAT(CAS) soil flooding (x100)



Soil before first flooding (x100)

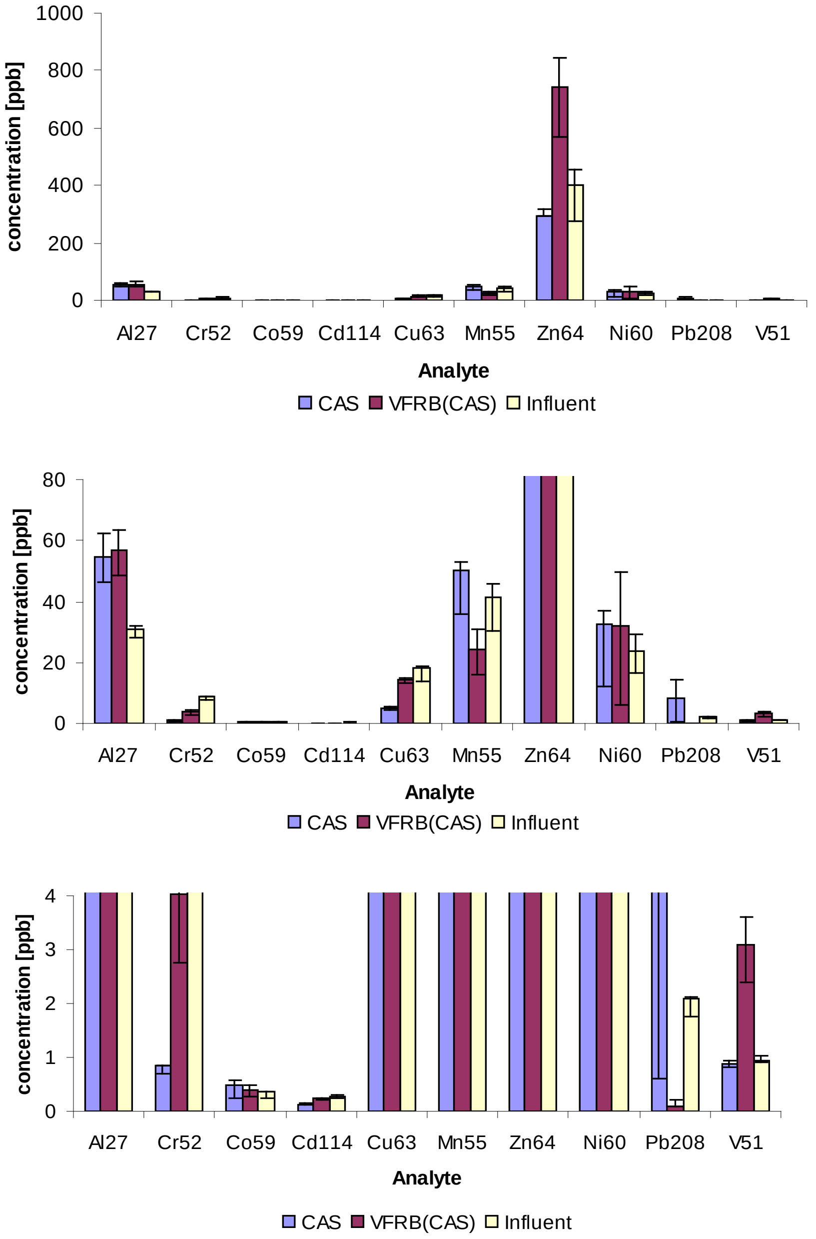


ESEM spectrum

APPENDIX IV Metal Analysis: Variability and Reproducibility

Variability:

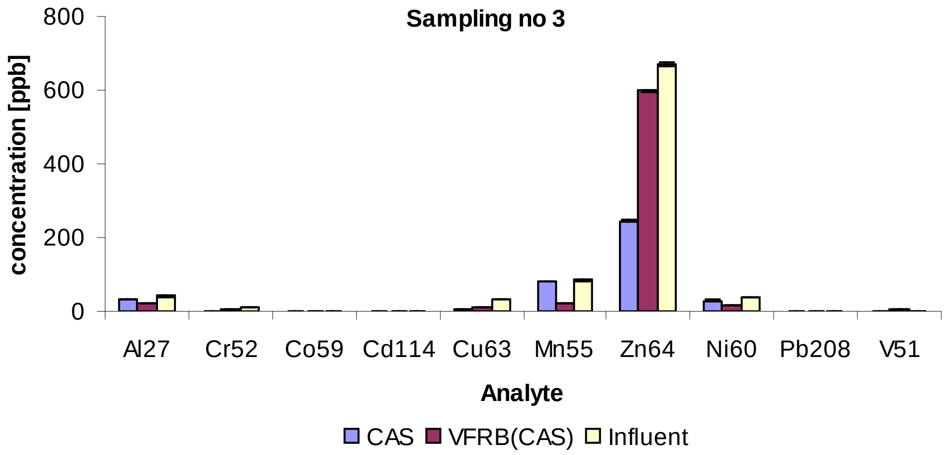
- 5 samples per effluent (CAS, VFRB(CAS) and influent) taken every two days at different times during the 12 days SRT period.
- Percentage error between 0.7 and 125% with average of 27.1 and 17.5% respectively for 0.25 and 0.75 centils.



Variability over time, at different scales.

Reproducibility

- 4 replicates per samples
- percentage error between 0.2 and 9.9% with an average of 1.7% for both quartils.



Reproducibility of ICP.

APPENDIX V Lettuces Trial

Material:

- 5 pots per type of feed
- 8 types of feed: tape water, fertiliser, and primary, CAS, MBR, VFRB(CAS), VFRB(MBR) and VFRB(primary) effluents.
- Lettuce seeds were planted in filter grade sand at 1g.cm^{-3}
- Temperature maintained at 15±2 °C
- Wetting of about 200 mL.(week.pot)$^{-1}$ (or 0.34 cm.d^{-1})
- Additional lightning using Philips TL-D 36w/35-535 lamps

Analyses:

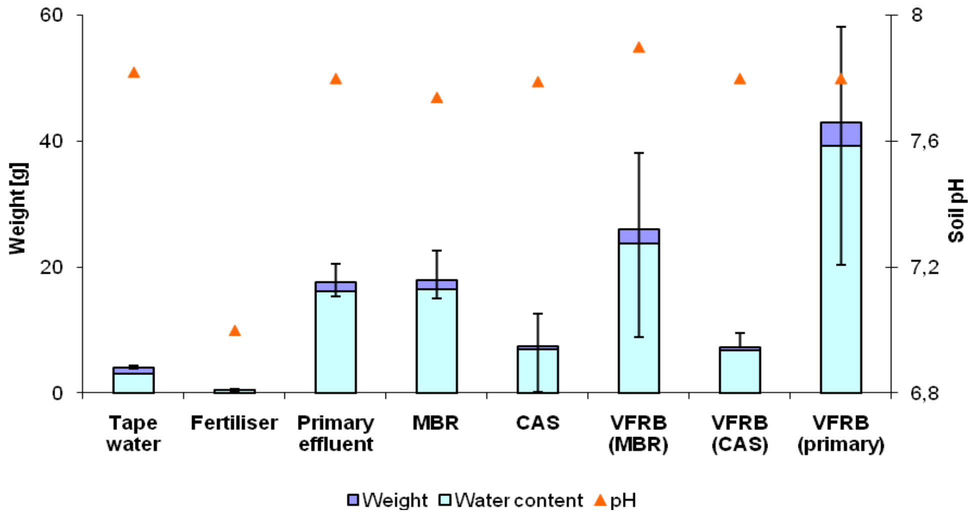
- Crop yield and water content (weight after 105 °C and 550 °C)
- Qualitative analysis of soil for ph, nitrogen, phosphorus, and potassium using Luster Leaf 1601 Rapitest Soil Test Kit

Pictures:

Greenhouse Dec. 2008

First sprouts (Dec. 2008) (left) and lettuces on sampling day (May 2009) (right)

Lettuce fed with fertiliser showing algae and fungi growth (right) compared to lettuce fed with CAS effluent (left) (sampling day, May 2009)



Results from lettuce trial

APPENDIX VI INTERACTIONS BETWEEN THE DIFFERENT ACTORS IN P AND N REMOVALS DURING SAT PILOT PLANT OPERATION