RECLAIM WATER

Water Reclamation Technologies for Safe Artificial Groundwater Recharge

Project no. 018309

Specific Targeted Research Project

Thematic Priority 'Global Change and Ecosystems'

Publishable Final Activity Report

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1 Summary description of project objectives

Solutions to global water stress problems are urgently needed yet must be sustainable, economical and safe. The utilisation of alternative water sources like reclaimed municipal wastewater is one of the most obvious and promising options in integrated water management. Among the various beneficial uses of reclaimed wastewater managed aquifer recharge (MAR)[1] receives growing attention because it features advantages such as additional natural treatment, storage capacity to buffer seasonal variations of supply and demand as well as mixing with natural water bodies which promotes the acceptance of further uses, particularly indirect potable use. Major concerns about the safety of this exploitation route of an alternative water source are connected to microbial and chemical contaminants occurring in wastewater, among which are emerging trace organics like endocrine disrupters and pharmaceuticals.

The strategic objective of this project is to develop hazard mitigation technologies for water reclamation providing safe and cost effective routes for managed aquifer recharge. The work assesses different treatment applications in terms of behaviour of key microbial and chemical contaminants. The knowledge generated in the project and the technologies developed are also suited to the needs of developing countries, which have a growing need of supplementation of freshwater resources. The project strategically supports the competitiveness of European technology suppliers and water services in the context of water reclamation and aquifer recharge.

The project focuses its research objectives on the investigation of water reclamation technologies and their impact on key contaminants. The project aims to provide new combinations of technologies to treat wastewater to appropriate water quality levels for sustainable groundwater recharge. RECLAIM WATER in its scope concentrates upon different issues indicated in Figure 1 following the water cycle in a water reclamation and aquifer recharge process:

1. Improvement of municipal wastewater treatment processes as an important prerequisite for water reclamation;

2. Optimised water reclamation technologies to produce feed water for aquifer recharge;

3. Design and operation of actual recharge systems (e.g. infiltration-percolation systems, constructed wetlands, lagoons, well injection);

4. The investigation of processes in the unsaturated soil zone and their dependence on the pre-treatment (steps 2 and 3);

5. The investigation and modelling of the introduction of a wastewater phase in a natural aquifer using tracer compounds (like Boron);

6. Anticipating the needs of various uses of recovered water (e.g. indirect potable use and non-potable uses) in terms of water quality requirements and definition of relevant target contaminants;

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[1] "Managed Aquifer Recharge" (MAR) comprises a large range of injection-recovery techniques. In the following we use "Managed Aquifer Recharge" instead of the formerly used artificial groundwater recharge as generic term to indicate that recharge of aquifers is not an artificial but well controlled measure to replenish depleted and overabstracted aquifers and provide (seasonal) storage.

7. Developing (if necessary) and applying the necessary analytical tools to monitor the relevant contaminants (microbial and chemical) including the investigation of contaminant fate in the processes;

8. Collecting existing data on the behaviour of the target compounds and the impact of treatment options in case studies to provide a sound basis for human health environmental risk assessment.
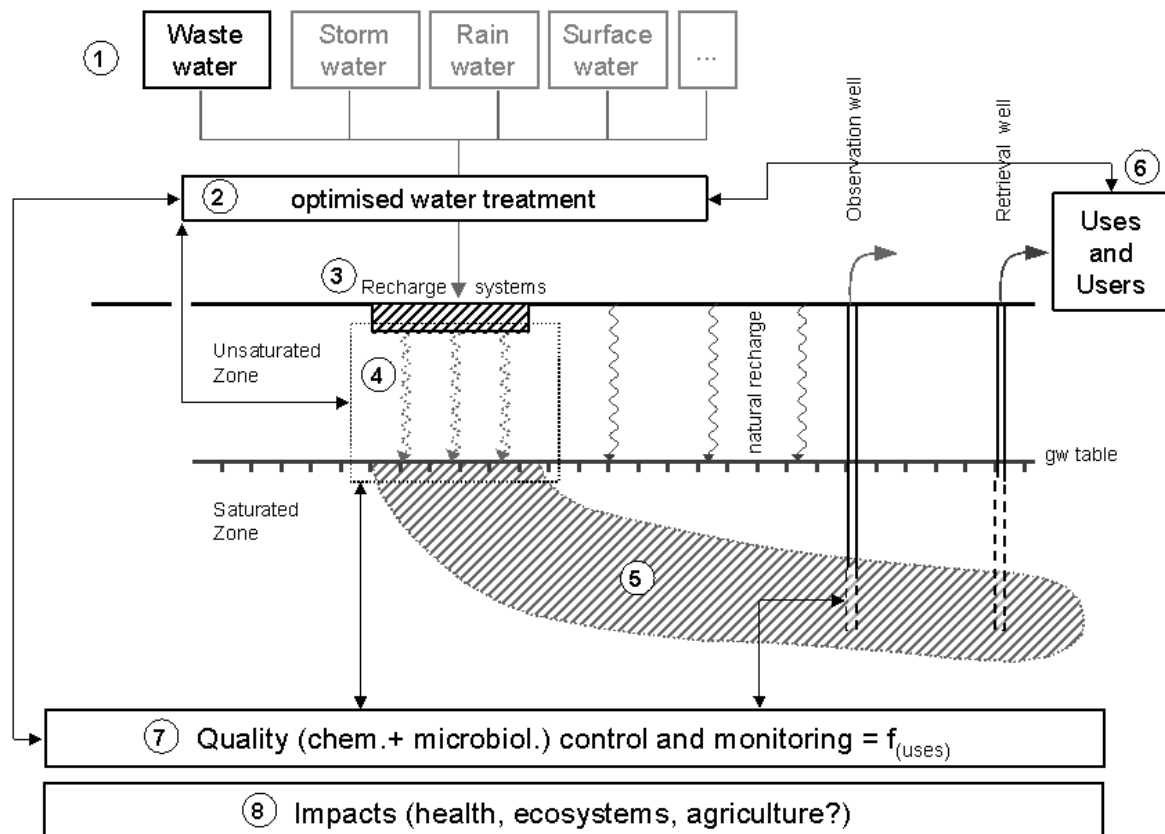

Figure 1.1 Concept of managed aquifer recharge and points of consideration

RECLAIM WATER integrates technological water reclamation solutions with natural attenuation processes occurring in the subsurface to achieve upgraded water quality assessed on the basis of key contaminants. The project directly relates the knowledge obtained on new treatment processes and contaminant behaviour to the question of risk associated to the indicated use. The risk studies cover water intake, treatment, storage and distribution steps, analytical tools, monitoring and control systems, and operational procedures as well as communication procedures. A coherent application of these elements in a number of case studies, that cover important reuse practices, result in recommendations all the way down to the end-user level, where risk management has to be practiced on a day-to-day basis. The project outcome contributes to the definition of "best available technologies" for wastewater reclamation and reuse applications in the sense of integrated technical, analytical and operational components.

WP1: Assessment and development of water reclamation technology

Water reclamation comprises a broad range of technologies from technical unit processes such as advanced oxidation processes, adsorption, and filtration applying porous and dense membranes to natural processes such as reed beds, wetlands, and soil passage. This work package has the objective to:

- Evaluate and optimise the cost effectiveness of the various alternatives for each single barrier

- Evaluate and optimise the cost effectiveness of the various alternatives for the entire treatment train based on a multi barrier approach

- Evaluate and optimise the cost effectiveness for the treatment of concentrates arising from dense membranes treatment (reverse osmosis and nanofiltration).

WP2: Analysis of microbial contaminants and antibiotic resistance genes

This workpackage adapts and develops new analytical methods to detect pathogens, antibiotic resistance genes and/or indicators and applies these methods to the set of case studies in WP4 and selected fate studies in WP5. These contaminants comprise:

- Six faecal contamination indicators

- Fifteen waterborne and emerging waterborne pathogenic micro-organisms (comprising viruses, protozoa, helminths and bacteria)

- Six antibiotic resistance genes

Furthermore the transport and the decay coefficients of microbial contaminants, important for the development of appropriate models and to predict their fate in natural environments have been studied by the diffusion cell technology in reed beds and recharged aquifers.

WP3: Analysis of organic compounds

The objectives of this workpackage were

(i) the development of analytical protocols for a list of more than 50 different trace organic compounds (antibiotics, disinfection by-products, industrial chemicals, iodinated contrast media, etc.) as well as for

(ii) bulk organic parameters such as NOM (natural organic matter) and EfOM (effluent organic matter).

(iii) application of these advanced analytical methods to the case study sites (WP4), selected pilot tests (WP1) and specific fate studies (WP5)

WP4: Technical water reclamation and aquifer recharge case studies

The primary objective of this workpackage is to provide an overall performance assessment of water reclamation for aquifer recharge in Europe and abroad. Specific goals of WP4 are to:

- implement and manage field testing protocols and timetables for each test site.

- facilitate the performance testing of novel treatment technologies and configurations developed at laboratory and pilot scale in WP1 and characterise their operational effectiveness, efficiency and feasibility at full scale.

- utilise the analytical techniques established under WP2 to characterise pathogen contamination in waters at various stages of the treatment process. Where treatment improvements are implemented at test sites, this characterisation will be conducted under both sets of conditions.

- utilise the analytical techniques established under WP3 to characterise organic pollutants (specifically antibiotic compounds) in waters at various stages of the treatment process. Where treatment improvements are implemented at test sites, this characterisation will be conducted under both sets of conditions.

- facilitate execution of the contaminant fate studies to be conducted under WP5 and the risk assessment and modelling work being performed under WP6 and WP7 respectively (primarily data provision).

WP5: Specific studies on removal and fate of microorganisms and organic compounds

The main objective of this workpackage is to elucidate the fate and the removal of pathogenic microorganisms, key organic contaminants and bulk organics in currently running water reclamation technologies including soil aquifer treatment during artificial recharge as well as more innovative technologies such as ozonation of effluents and brine.

- Fate and formation of tracer substances and toxic disinfection by-products during water reclamation

- Fate of bulk organics and specific contaminants after ozonation and subsequent soil passage

- Fate and behaviour of specific organic pollutants in Soil-Aquifer-Treatment

- Fate of microorganisms in Soil-Aquifer Treatment and ASTR

- Removal of contaminants in brine treatment of water reclamation installations

WP6: Case-study related risk studies

This workpackage has the objective to apply several up-to-date risk assessment and risk management tools to the case studies. Tested approaches comprise:

- Case study specific risk assessment and hazard analysis

- Case study specific hazard HACCP plans

- Development of recommendations on HACCP applications

A risk communication tool for recycled water systems is also provided.

WP7: Integrative monitoring, modelling and assessment tools for subsurface treatment and storage

The overall objective of research activities within this workpackage is to apply hydrogeological and biogeochemical modelling as well as monitoring:

- to provide an improved understanding and novel tools for the prediction of the transport and fate of organic and inorganic pollutant induced particularly by SAT and

- to extend and validate existing models for ASTR .

The research is based on an experimental and numerical approach that integrates the dynamics of chemicals and microbes in subsurface environments (saturated and unsaturated zones), with novel biogeochemical models and environmental tracers. The use of coupled flow and geochemical modelling already demonstrated and explained the underperformance of recharge schemes due to the specific conditions prevailing in the underground environment.

The ultimate goal of this effort is to obtain new insights on coupled hydrological, geochemical, and microbial processes that influence contaminant migration and integrate these experimental findings with an advanced software system calibrated on the lab and pilot scale and validated on the field scale.

2 Project consortium and coordination

The multi-national and interdisciplinary consortium carrying out the project is given in the Table 1 below:

Table 2.1 The RECLAIM WATER Consortium

Partic. Role*	Partic. no.	Participant name	Participant short name	Country	Date enter project	Date exit project
CO	1	Rheinisch Westfälische Technische Hochschule Aachen	RWTH	Germany	1	39
CR	2	Consiglio Nazionale delle Ricerche	IRSA	Italy	1	39
CR	3	Technische Universität Berlin	TUB	Germany	1	39
CR	4	Swiss Federal Institute for Environmental Science and Technology	EAWAG	Switzerland	1	39
CR	5	Cranfield University – School of Water Science	CRAN	United Kingdom	1	39
CR	6	University of Barcelona	UB	Spain	1	39
CR	7	DHI Water & Environment	DHI	Denmark	1	39
CR	8	Institute for Ecological Engineering	IEI	Slovenia	1	39
CR	9	Ribo Technologies BV	MICRO	Netherlands	1	12
CR	10	Mekorot Water Company	MEK	Israel	1	39
CR	11	Unesco IHE	IHE	Netherlands	1	39
CR	12	Federal Institute of Hydrology	BFG	Germany	1	39
CR	13	Tsinghua University	INET	China	1	39
CR	14	Bureau de Recherches Geologiques et Minieres	BRGM	France	1	39
CR	15	Aquafin NV	AFIN	Belgium	1	39
CR	16	United Water	UW	Australia	1	39
CR	17	National Autonomous University of Mexico - Institute of Engineering	UNAM	Mexico	16	39
CR	18	The Council for Scientific and Industrial Research	CSIR	South Africa	16	39
CR	19	Public Utilities Board	PUB	Singapore	16	39
CR	20	National University of Singapore - Centre for Water Research	NUS	Singapore	16	39

*CO = Coordinator   CR = Contractor

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3 Methodologies, approaches employed and results

The work carried out in the project comprised research, innovation-related and development as well as management activities. The activities were structured in well connected work packages (WPs) as illustrated in Figure 3.1. The methodology as well as the results and innovation of the activities are described subsequently.
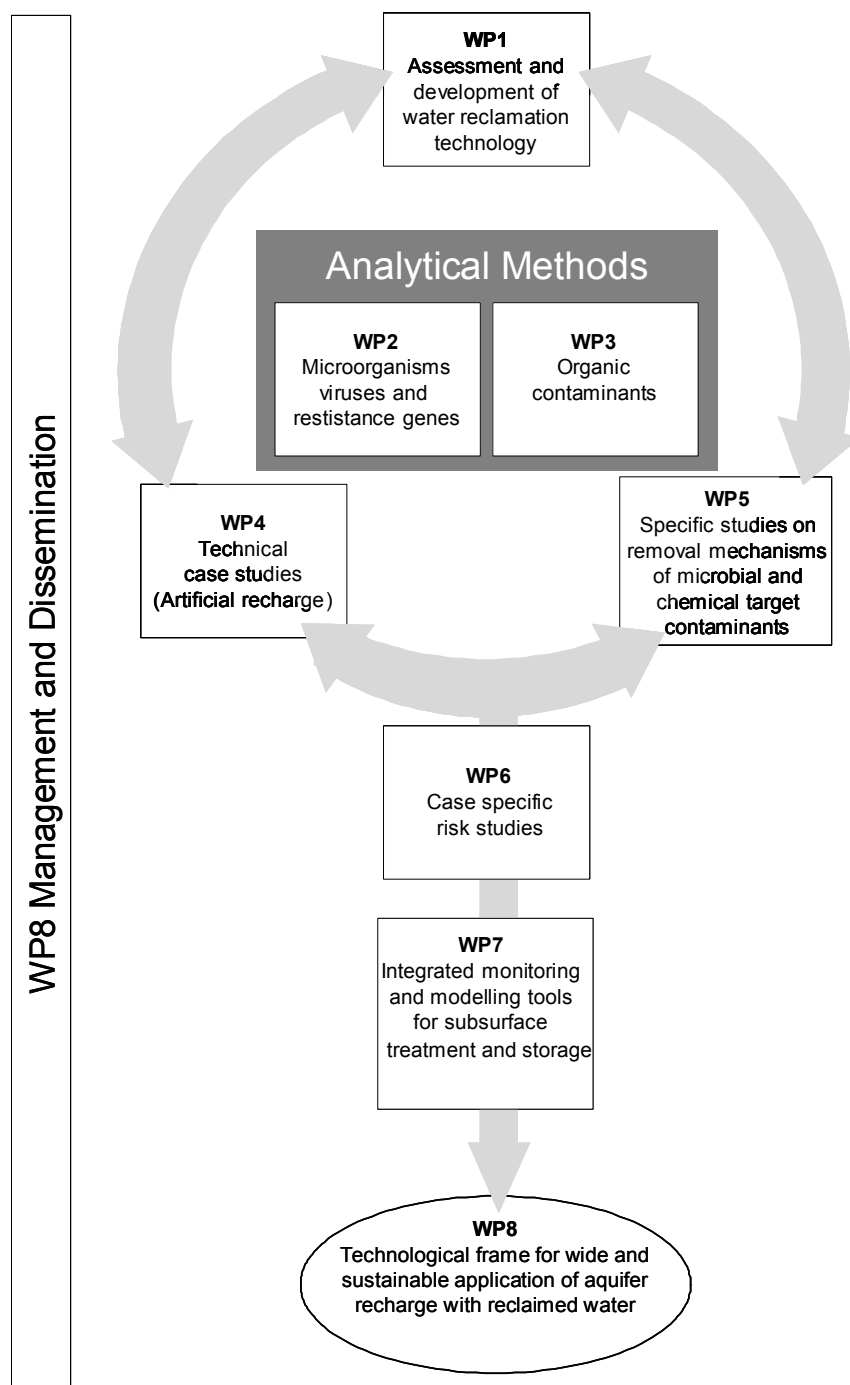

Figure 3.1 The RECLAIM WATER work packages and their interconnection

RECLAIM WATER Case Studies

To provide a better understanding of aquifer recharge, from 2005 to 2008, eight international pilot or full scale test sites treating municipal wastewater effluent and/or stormwater for aquifer recharge have taken part in WP4 activities (cf. Table 2 and Figure 3.6) which included the monitoring of more than 50 basic wastewater parameters and contaminants at different location of their schemes and at least three times per year, except for the partners who joined the project in its second year (*i.e.* South Africa and Mexico).

The main objective of this study was to assess the overall performance of these sites in recharging aquifers mainly for irrigation and potable water supply purposes by following contaminant fate throughout each scheme. The reclamation technologies used at the sites covered a range of pre-treatment technologies from conventional activated sludge to membrane technologies, and a range of recharge methods including river bed filtration, dune filtration, sinkhole infiltration, infiltration ponds, soil aquifer treatment and well injection. This was an opportunity to compare different recharge methods under various geological and pre-treatment conditions, hence providing a broader knowledge of managed aquifer recharge for sustainable groundwater recharge in both developing and developed countries.
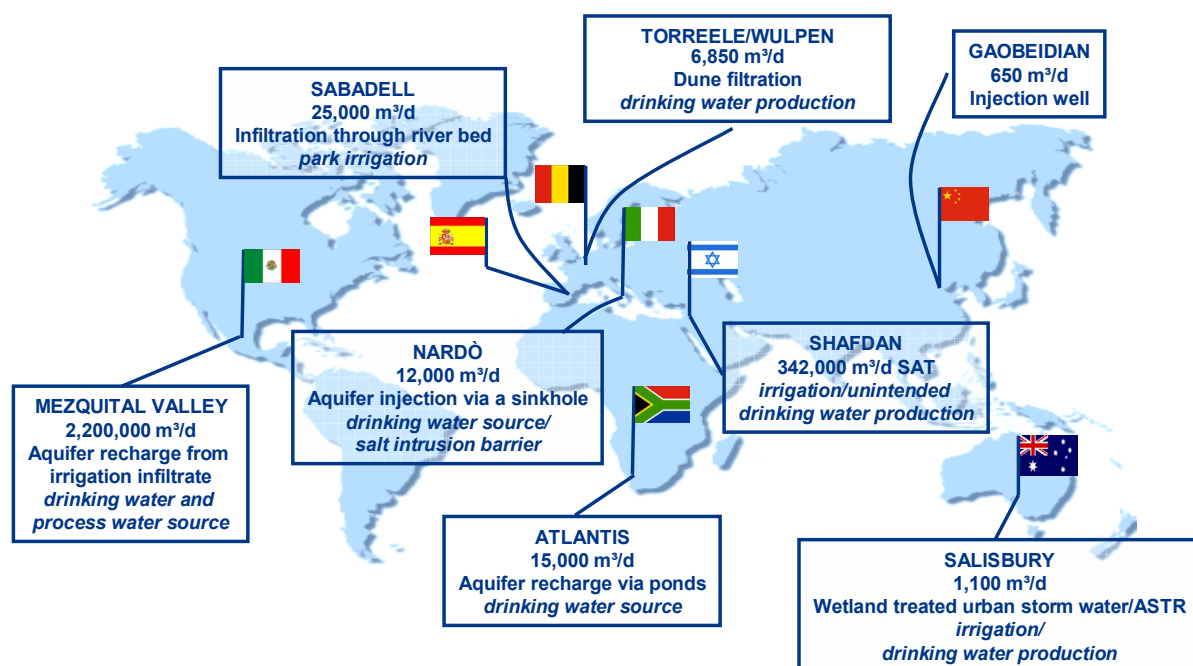

Figure 3.2 RECLAIM WATER case studies of MAR

Table 3.1 Characteristics of the eight RECLAIM WATER case study sites

Test Site	Injected source	Advanced water treatment	Recharge method	Post treatment	Recharge rate	Reuse purpose
Sabadell SPAIN	Secondary effluents (activated sludge + nutrient removal)	none	Infiltration through river bed	UV + Chlorination	19000 m^3/d (2006)	Public park irrigation Street cleaning
Nardó ITALY	Secondary effluents (conventional activated sludge + biological treatment plant)	chlorination	Injection via a sinkhole	-	4.4 Mm^3/y (average)	Salt intrusion barrier + drinking water source
Shafdan ISRAEL	Secondary effluents (activated sludge + nutrient removal incl. Bio-P)	a) full scale: none b) pilot unit: Ultrafiltration	Soil Aquifer Treatment a) infiltration ponds b) dug well	Intermediate chlorination against biofouling in SAT conveyance pipe	339000 m^3/d (full site) ~120 m^3/d (pilot site)	Irrigation (unintended drinking water quality)
Gaobeidian CHINA	Secondary effluents (activated sludge + nutrient removal)	coagulation + sand filtration + ozonation + slow sand filter	Well injection	-	350-500 m^3/d	*None (pilot site)*
Torreele/ Wulpen BELGIUM	Tertiary effluents (low loaded pre-denitrification activated sludge system + simultaneous chemical P-removal)	Ultafiltration + chlorination + reverse osmosis	Dune infiltration	Chlorination, aeration, rapid sand filtration + UV disinfection prior to distribution	2.5 Mm^3/y	Sustainable groundwater management Potable water supply
Salisbury AUSTRALIA	Urban stormwater	Wetlands (in-stream basins + holding storage basins + cleansing wetland)	Aquifer storage transfer and recovery (ASTR)	-	11 L/s (ASTR)	Intended irrigation, industrial use and drinking water production
Tula Valley MEXICO	Raw wastewater mixed with stormwater and natural surface water	none	Irrigation (atypical SAT)	Chlorination (only for drinking water production)	25 m^3/s	Industrial use. Domestic use. Potable water production. Irrigation
Atlantis SOUTH AFRICA	Secondary effluent (nitrification-denitrification steps (anaerobic-anoxic-aerobic) mixed with Urban stormwater runoffs	none	Infiltration ponds	Ion exchange + Chlorination	2.7 $Mm³$/y	Potable water supply

WP1: Assessment and development of water reclamation technology

BACKGROUND

The experimental work focused on polishing options for upgrading wastewater after physical or biological treatment (i.e. primary or secondary effluent) for groundwater recharge. Technologies based on the following treatment principles were studied:

a) Naturally based treatments as soil passage, soil aquifer treatment (SAT), wetland treatment and reed bed,

b) Chemical oxidation with ozone,

c) Membrane separation based on porous membranes (microfiltration, ultrafiltration, membrane bioreactor) or on dense membranes (nanofiltration or reverse osmosis) and options for treating brines originating from membrane separation,

d) Physico-chemical separation by sorption either on activated carbon as sorption medium or via capacitive deionisation.
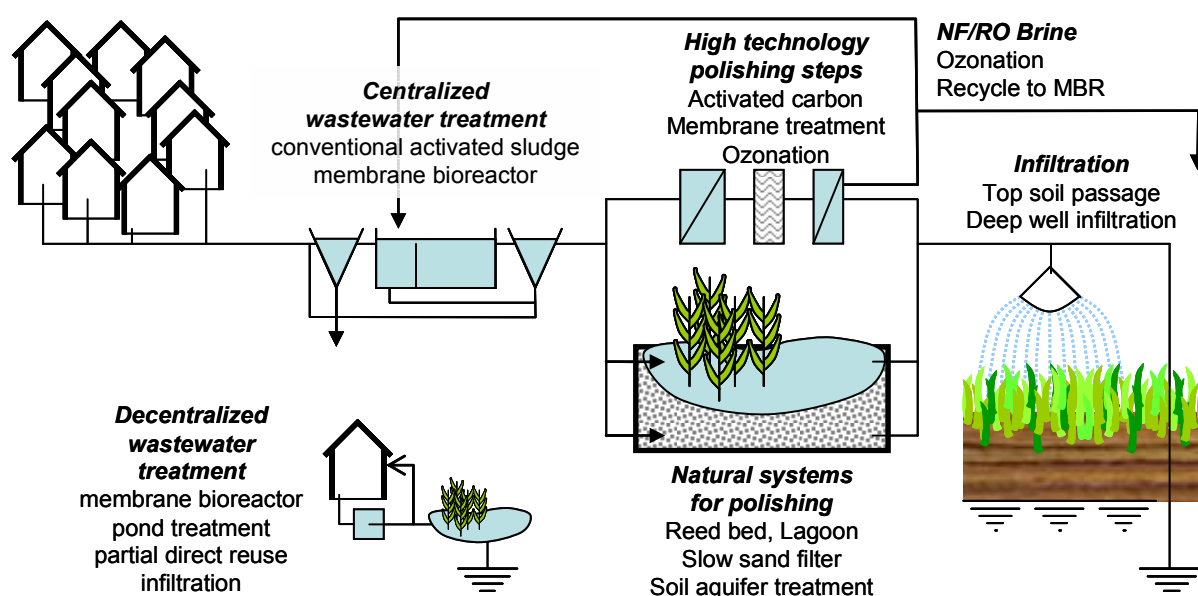

Figure 3.3 Schematic summary of the treatment steps investigated on within WP1

METHODOLOGY AND RESULTS

Soil passage, soil aquifer treatment (SAT) and reed bed

Soil column experiments have been performed to assess the fate of bulk effluent organic matter (EfOM) in soil column experiments simulating soil passage. Results derived from fluorescence excitation emission matrix (F-EEM) and liquid chromatography with organic carbon detection (LC-OCD) indicate preferential removals of protein-like and polysaccharide-like organic matter over humic-like organic matter. This result has implications for post-treatment by membrane filtration (fouling reduction) and chlorination (disinfection by-products), and as a carbon source(s) for driving heterotrophic biodegradation of organic micropollutants. Further nitrification as well as denitrification has been shown to occur during

soil passage. With respect to DOC removal SAT is equally effective in treatment of primary effluent from wastewater treatment plants as in the case of treatment of secondary effluent, when adequate depth (travel time) is provided. DOC removal is comparable, if the soil is aerobic or anoxic. At the Shafdan site, 6 to 12 months SAT treatment resulted in DOC values below 1 mgDOC/L.

Primary effluent after coagulation with 40 mgFe^{3+}/L achieved comparable DOC values after soil passage as secondary effluent after soil passage (i.e. 15 and 10 mgDOC/L respectively).

A lab scale soil aquifer treatment setup simulating karstic soil removed 3 to 5 log units of several microbial indicator parameters (i.e. total bacterial count, faecal coliforms, enterococci, *clostridium* spores and somatic coliphages). Similar removal was observed in the karstic reference site at Nardò, Italy.

SAT was shown to partly remove micropollutants with better removal rates at higher residence times (6 – 12 months).

Concerning nutrient removal (N and P), suspended solids and turbidity, reed bed post-treatment yielded comparable quality if fed with wastewater treated with a conventional nutrient removal plant or a membrane bioreactor. Only if fed with primary effluent the ammonia concentrations significantly increased. In the case of MBR effluent the reed bed post treatment significantly increased the suspended solids and the bacterial counts, resulting in similar values as for reed beds fed with conventional activated sludge (CAS) treatment effluent. Activated sludge treatment combined with reed bed treatment resulted in 80 to 90% removal for several micropollutants studied (carbamazepine, diclofenac, ibuprofen, naproxen, galaxolide and tonalide).

Ozonation

Effluent upgrading with typical doses of 0.6 - 0.7 mgO$_3$/mgDOC are expected to remove most of the organic micropollutants. The work within Reclaim showed, that while ozonation itself did little change on the DOC concentration measured in the effluent, it oxidized aromatic structures (reduction in UV absorption as measured by LC-UVD) resulting in better degradability of the DOC in the following soil passage.

Membrane separation

Porous membranes (microfiltration, ultrafiltration and membrane bioreactors) remove pathogens, suspended solids and polymers (proteins and polysaccarides). All other quality parameters (including nutrients and organic micropollutants) may depend significantly from the design of the preceding biological treatment, but not on the porous membrane separation. For post-treatment with soil passage the removal of particulates and polymers may impact on soil clogging, oxygen uptake rate and denitrification capacity of the soil.

Additionally to the removal capability of porous membranes, dense membranes are able to retain also lower molecular compounds and ions to an extent strongly dependent on the specific membrane product used. Main governing factor is the molecular weight cut-off (MWCO) of the membrane, but also process conditions and compound properties influence the specific removal. Some compounds sorbing at dense membranes, e.g. Bisphenol A, can pass the membrane and especially compounds close to the MWCO or below it are only poorly retained by dense membranes. Nitrosamines such as NDMA and benzotriazole are not fully retained by reverse osmosis membranes. By rejecting ions, dense membranes retain also salinity and most heavy metals.

Finally as significant drawback, dense membranes produce a concentrate containing all the retained compounds requiring either further treatment or disposal. Brine treatment was tested

with capacitive deionisation (CDI), GAC/MF, ozonation and reed beds. Piloting CDI confirmed its capacity to remove ions to a degree allowing to recycle the treated brines back to the dual membrane treatment and thus. increasing the total water yield from 75% to >95%. The CDI featured significantly less fouling if preceded by an ultrafiltration removing all particulates. The combination of GAC/MF allowed for significant removal of organic micropollutants and pathogens, while total nitrogen as well as suspended solids were only slightly improved (total nitrogen <20%, SS 25%). Brine treatment with reed bed removed mainly the suspended solids (up to 90%) and part of the total nitrogen (40%). Both parameters are of relevance for the brine discharge tax at the investigated dual membrane reclamation plant in Wulpen/Torreele (Belgium).

Combination of membranes and activated carbon

Following the multi-barrier principle, processes can be combined to increase the removal capacity and the robustness of a reclamation plant. Different options of combining activated carbon and membranes have been tested: a) using granular or powdered activated carbon (GAC or PAC), b) using porous membranes or nanofiltration, c) placing the membrane step before or after the activated carbon treatment. Activated carbon mostly served as physical sorption media, although in case of GAC with long permanence times biological activity may also develop. The role of the porous membrane is to retain PAC and particulates. Combining nanofiltration with activated carbon is seen as a multi-barrier solution for micropollutants, and in case of GAC also for particulates and pathogens since the retention is based on independent mechanisms. The micropollutant retention strongly depends on the exchange rate of the carbon (50 mgPAC/L were needed for removing most micropollutants by >80 to 99%) and on the type of nanofiltration membrane. Some process combinations such as the PAC/NF hybrid process requires specific membranes, i.e. capillary nanofiltration, which are able to cope with higher particulate concentrations in the feed.

Costs and feasibility

Since most processes studied are still at a piloting stage, cost and feasibility can currently only be grossly estimated. For membrane based systems total costs and energy are estimated in the range of 0.25 to 0.50 €/m³ of product water and 0.3 and 1 kWh/m³. Combined activated carbon and membrane systems are estimated in 0.40 - 0.80 €/m³ and 0.5 - 1 kWh/m³.

No cost estimation is available for soil passage, SAT or reed bed treatment.

CONCLUSIONS

Several technical options for upgrading water quality prior to groundwater recharge are currently available consuming less than 1 €/m³ and 1 kWh/m³. Since comparative full scale performance is not yet available, a definitive ranking of technologies tested in pilot scale is currently not possible.

Natural systems (e.g. soil passage and SAT) clearly show a great upgrading potential allowing lower treatment costs and no residual streams compared with membrane systems. The final product water quality contains higher organic concentrations if compared with high pressure membrane applications. Concerning some micropollutants natural systems can only achieve a partial removal.

WP2: Analysis of microbial contaminants and antibiotic resistance genes

BACKGROUND

Determination of the microbiological water quality

A main concern in wastewater reuse is the microbiological quality of water and the possibility of spreading diseases. This workpackage aimed at developing new molecular quantitative detection methods for microbial water contaminants and evaluate the microbial quality of the water in three reclamation case study sites. Different microbial parameters were investigated:

a) Helminth eggs (*Ascaris lumbricoides, Trichurus trichiura, Ancylostoma duodenale*

b) Protozoa (*Gardia spp, Cryptosporidium spp*)

c) Viruses (Enterovirus-*Poliovirus, Echovirus, Coxsackievirus*-, *Hepatitis A virus* (HAV) and *Norovirus*.genomic group I and II)

d) Bacteria (*Campylobacter* spp*., Yersinia enterocolitica, Helicobacter pylori, Mycobacterium avium, Salmonella* spp).

e) Antibiotic resistance genes (*ampC*-ampicillin resistance, *mecA*-methicillin resistance, *SHV*-extended β-lactam resistance, *ermB*-erythromycin resistance, *tet*(O)-tetracycline resistance, *vanA*-vancomycin resistance)

Case Study Sites

Three sites utilised for indirect potable or irrigation reuse have been sampled for one year to monitor microbial water contaminants qualitatively and quantitatively:

1) Sabadell (Spain): At this site treated domestic wastewater is discharged into the Ripoll River and infiltrates into groundwater by bank filtration. The product water is used for irrigation of public parks after UV and Cl_2 treatment. Due to the vicinity of the park to hospital and the applied spray irrigation, pathogenic contamination of the irrigation water is a major concern (cf. Table 3.1).

2) Nardò (Italy), where treated domestic wastewater is transported in an open channel and injected by a sinkhole into a karstic aquifer. The water is withdrawn at a distance of 3 km from the sinkhole for irrigation. This case study served to study the pathogen transport and decay in a karstic aquifer (cf. Table 3.1).

3) Wulpen/Torreele (Belgium): This case study exemplifies the most advanced treatment train including ultrafiltration and reverse osmosis prior to infiltration ponds. The product water is withdrawn and distributed after further treatment for drinking water supplies (cf. Table 3.1).

In particular the presence of 15 waterborne pathogens, six antibiotic resistance genes and six indicators of microbial contamination was monitored in three to four campaigns during one year with five to six sampling points at each site.

Pathogen decay studies

This workpackage also focused on the study of decay coefficients of pathogens and indicator microorganisms. The effectiveness of diffusion cell technology in quantifying pathogen attenuation rates was evaluated and applied in reed beds used as pre-treatment to Aquifer Storage Transfer and Recovery (ASTR) and in recharged aquifer.

RESULTS

New detection methods and protocols

Protocols for sampling, sample concentration, storage and delivery of waters with different degree of pollution were developed and/or selected for all the microbial contaminants monitored at the three case study sites.

New quantitative detection methods, by real time PCR, were developed for six antibiotic resistance genes, four pathogenic bacteria and two viruses. The new methods were successfully employed on environmental water samples in WP4.

A Peptidic Nucleic Acid Fluorescence In Situ Hybridization (PNA FISH) protocol was adapted for the determination of the human pathogenic species of *Campylobacter* (*C. coli, C. lari* and *C. jejuni*) in environmental waters samples (Figure 3.3). The used molecular approach allowed a rapid and specific identification of this emerging waterborne pathogen and gave the same results as the complex and time consuming traditional cultivation method.
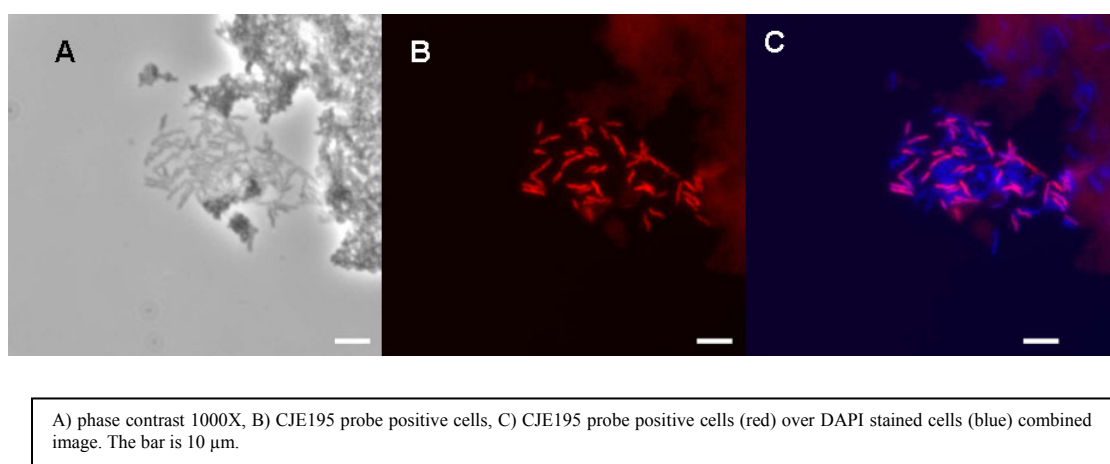

A) phase contrast 1000X, B) CJE195 probe positive cells, C) CJE195 probe positive cells (red) over DAPI stained cells (blue) combined image. The bar is 10 μm.

Figure 3.4 Campylobacter cells detected by PNAFISH in a Nardò enriched water sample

Monitoring of the sites

Sixty samples from the three European Sites, in three to four sampling campaigns, have been analysed. The main results can be summarised as follows:

Antibiotic resistance genes: The three aquifer recharge systems demonstrated different removal capacity for antibiotic resistance genes. Ultrafiltration and reverse osmosis in the Wulpen plant proved to be very efficient barriers for the elimination of antibiotic resistance genes. On the contrary SAT followed by UV treatment and chlorination in Sabadell and, in Nardò, the fractured and permeable aquifer only partially removed these contaminants. In these two artificial recharge sites it was demonstrated that *tetO, ermB* and *mecA* can occur in groundwater suggesting that groundwater may be a potential source of antibiotic resistance in the food chain (Bockelman *et al* 2009).
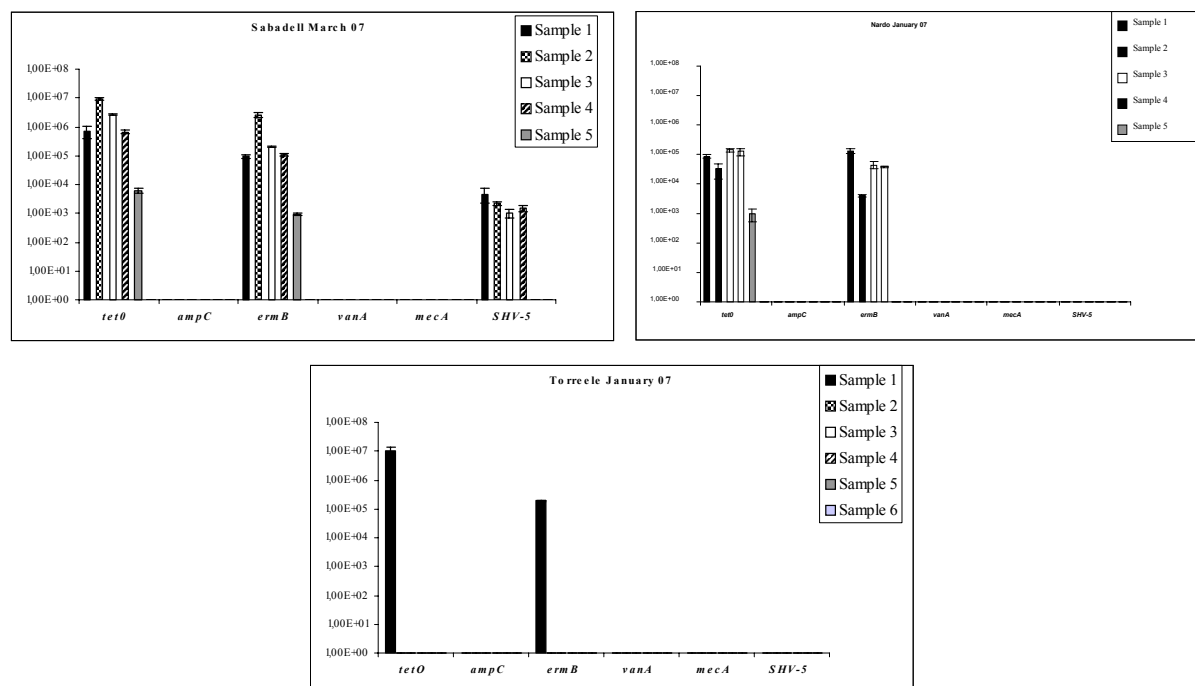
Figure 3.5 Elimination efficiency for antibiotic resistance genes at the artificial groundwater recharge sites (Bockelman et al 2009)

Protozoa: *Giardia* and *Cryptosporidium* were frequently detected in waters of low bacteriological quality (WWTP effluents and surface waters) indicating that these could be a potential source of human infections. MAR systems in Wulpen and Sabadell proved to be effective in the removal of these parasites which were completely eliminated from the final water product. Also in Nardò there was an evident decrease of cysts and oocysts during the underground flow but water withdrawal at 500m distance () from the aquifer recharge point was not depleted of these parasites.

Viruses: No viruses were detected either in Sabadell and Wulpen. In Nardò HAV was never found, while Enteroviruses were found by RT-PCR only in 8 samples out of 37, and none of them could infect cell lines (BGM, Buffalo Green Monkey), indicating that infectivity had been lost. Norovirus (GGI and GGII) were found in the February 2007 campaign; Norovirus II presented decreased values from the WWTP effluent, sinkhole, well at 300 m and well at 500 m (2,000, 1,000, 20, and below detection limit, genomic copies (gc)/L). These data are quite important to model the natural decays of these pathogens.

Salmonella spp.: *Salmonella* spp were never detected in samples from Wulpen. In Nardò *Salmonella* spp. were detected in two individual samples (once in WWTP effluent and once in growndwater) in two campaigns out of four at a level of 10^3 gc/100mL. The presence of *Salmonella* spp. is particular relevant at Nardò site, because there is a stringent limit for *Salmonella* presence in reclaimed water in Italy: they have to be absent in 100 ml (D.M. 185 2003). In Sabadell *Salmonella* spp. were found only in one sampling campaign March 2008, in all the five samples.

Campylobacter spp. These bacteria were detected only twice, once in Nardò and once in Sabadell site. In both cases they were detected in surface water samples, directly influenced by effluent and surface water run-off, before SAT.

<u>Helminth eggs</u>: They were found in very limited number mainly WWTP effluent and never in the final sampling points (MAR product water).

Yersinia enterocolitica, Helicobacter pylori, Mycobacterium avium have never been found in any of the samples analysed in the three case study sites, apart from a weak positive sample for *Yersinia enterocolitica* found in the WWTP effluent of Nardò.

Water quality after MAR treatment

None of the microbial contaminants investigated was found after the treatments in Wulpen.

In Sabadell the aquifer barrier is sufficient to remove pathogenic organisms, even though there were all samples from the March 2008 sampling campaign positive for *Salmonella*, it has to be stressed that the presence of *Salmonella* spp. is determined and expressed in terms of genomic copies, and this number also includes dead bacteria.

In Nardò several contaminants were found, but it was possible to observe that there was a natural depuration capacity of the karstic system, with a decrease of viruses and protozoa, during the ground water passage. In Nardò the well at 500 m resulted positive twice for *Salmonella.*. The Soil Aquifer Treatment in Nardò shows a certain level of natural water disinfection (La Mantia et al. 2008), evidently lower than that observed in the other sites. The Nardò site, requires the adoption of a longer withdrawal distance, for caption of water in safe conditions.

Pathogen attenuation study

Diffusion chambers have been used to study the decay rate of selected enteric microorganisms in captured urban stormwater within a constructed reedbed used as a pre-treatment step prior to an Aquifer Storage, Transfer and Recovery (ASTR) scheme and in the aquifer[2].
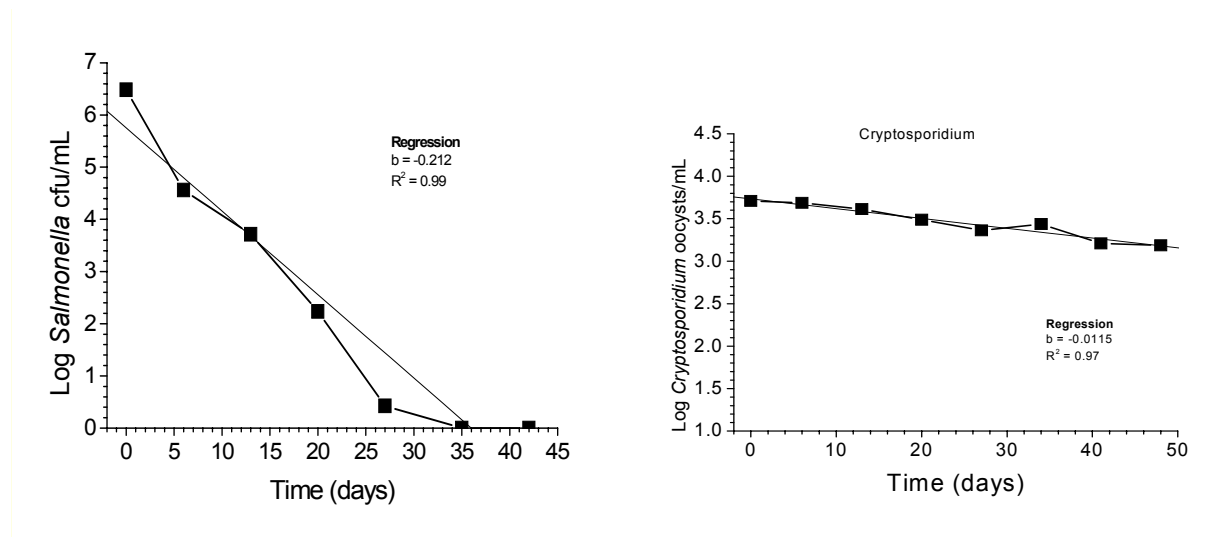

Figure 3.6 Decay slopes and corresponding regression lines for S. typhimurium cells and Cryptosporidium parvum oocysts in decay chambers

[2] Toze *et al*. (2008) Decay of Microbial Pathogens in a Constructed Reedbeds Receiving Storm Water for Pre-Treatment Prior to Aquifer Storage and Recovery. CSIRO: Water for a Healthy Country National Research Flagship; Toze *et al.*(2009) Decay of Enteric Pathogens in Urban Stormwater Recharged to an Aquifer using Aquifer Storage, Transfer and Recovery; A report to the ASTR Research Board. CSIRO: Water for a Healthy Country National Research Flagship

The microorganisms studied were *E. coli, Enterococcus faecalis, Salmonella typhimurium*, adenovirus and *Cryptosporidium parvum* oocysts for the reedbed study while in the aquifer also *Campylobacter jejuni*, Coxackievirus and Rotavirus were analyzed. In Figure 3.5 the decay of *Salmonella typhimurium* and *Cryptosporidium* oocysts in reedbeds are shown as an example.

The decay rates for the studied microorganisms in both the reedbed and the ASTR system are given in Table 3.2.

Table 3.2 Decay times for a 90% loss (T90) of pathogens and indicator microorganisms in groundwater at the ASTR site.

Pathogen/Indicator	T_{90} (days)	
	Reedbed	ASTR
E. coli	4.3	0.1
Enterococcus faecalis	6.2	2.5
Salmonella typhimurium	4.7	0.7
Campylobacter jejuni	ND[1]	0.2
Coxsackievirus	ND	109
Adenovirus	32.6	59
Rotavirus	ND	185
Cryptosporidium parvum oocysts	87	86

[1] ND = Not done.

An assessment of the ability of pathogens to be removed by natural processes in the aquifer has demonstrated that there were good removal efficiencies for all of the bacteria. The removal times for *Cryptosporidium* oocysts were consistent in both reedbed and the aquifer. The decay of the viruses was more varied with different removal times in the reedbed and the aquifer with rotavirus in the aquifer having the slowest removal times of all the microorganisms studied in both the aquifer and the reedbed.

CONCLUSIONS

The monitoring of waters hygienic quality in MAR systems is at present based on the detection of microbial indicators and the use of standardised protocols/methods developed for drinking water. Within WP2, methods for sampling, concentration and quantitative detection of microbial water contaminants in MAR systems were developed and applied, producing a important set of data for the future investigation of aquifer recharge schemes.

Furthermore the knowledge, important for operation and planning of MAR schemes, on the prevalence and distribution of antibiotic resistance genes and pathogenic microorganims and on their persistence in the water environment (decay rate results) was increased.

WP2 results showed that advanced water treatment technology with ultrafiltration and reverse osmosis (Wulpen/Torreele) produces water of drinking quality fully depleted of pathogens. It was proven that reuse of well treated wastewater is not a source of diffusion of antibiotic resistance gene in the environment.

Different efficiencies of pathogens and antibiotic resistance genes removal were observed by natural attenuation systems at Sabadell (SAT) and Nardò (water flow in the karstic fractured subsoil) recharge sites. The results contribute to the comprehension of limits and potentialities of low costs treatments for MAR scheme that are of major importance in developing countries.

WP3: Analysis of organic compounds

According to the tasks of WP3 - comprising the development of analytical protocols for (i) a list of more than 50 different trace organic compounds (antibiotics, disinfection by-products, industrial chemicals, iodinated contrast media, etc.) as well as for (ii) bulk organic parameters such as NOM (natural organic matter) and EfOM (effluent organic matter) -, respective analytical methods have been adapted and developed within the first year of the project (Deliverables 3.1 and 3.2). During the 2nd and 3rd year, this workpackage coordinated the organic micropollutant sampling programs and sampled (iii) across five of the eight case study sites in Israel, Belgium, Italy, Spain and China. Each site was sampled in at least three campaigns for considering seasonal changes. The newly developed methods ('protocol 2') were applied at least once at each location, with the exception of those sites where the measurement of certain parameters was not sensible (e.g. DBP in China, where no disinfection is applied). A comprehensive summary (iii) of individual site results on protocol 2 was given towards the end of the second year (Milestone 3.2) with a final update half a year later. Next to the development of analytical methods, sampling coordination, sampling and measurement at the selected five case study sites, WP3 performed service analysis for WP1 and WP5 partner (iv). Moreover some analytical campaigns were conducted for the TTC partners from South Africa and Mexico (v).

The investigated demonstration sites are characterised by different treatment process trains, covering a wide range of wastewater treatment processes. The following technical pathways were investigated:

A) Treated domestic wastewater injected by a sink hole into a fractured and karstic aquifer with relatively low residence time (< 30 days) (Nardó, Italy).

B) Treated domestic and industrial wastewater discharged into a river and infiltrated by bank filtration into the groundwater (water is recycled for lawn irrigation) (Sabadell, Spain).

C) Treated domestic wastewater treated by a low pressure membrane filtration (ultrafiltration, UF) prior to infiltration into a dug well with short term soil aquifer treatment (SAT retention time < 2 months) (Pilot unit, Shafdan) and

D) at the same location and source water quality infiltration for long term SAT with 6 – 12 months of retention time (Shafdan full scale plant).

E) At location (E) treated wastewater is processed in an integrated membrane system (UF followed by reverse osmosis) prior to pond infiltration and mid term SAT (Wulpen/Toreele Plant)

F) Domestic wastewater, coagulated (polyaluminum chloride) and ozonated before percolation for short term SAT treatment (Gaobeidian plant).

The comparison summarizes the variety of organic water constituents in secondary effluents, advanced treated waters and finally abstracted ground water. The fate and behaviour of refractory organic compounds during different treatment steps and different SAT retention is

of specific interest. For example carbamazepine was found in all secondary effluents, except from one (where the antiepileptic is obviously not prescribed), varying from 60 – 1400 ng/L. Carbamazepine was also found in effluent recharged groundwaters but at much lower concentration (10 – 600 ng/L). Due to its high mobility and non-biodegradability carbamazepine is therefore identified a wastewater indicator, the same situation exists for diatrizoate which was found in several samples in the lower µg/L range.

In general iodinated contrast media (ICM) contribute to the highest share of single substances in domestic effluents as well as in recharged groundwater. Especially diatrizoate, iohexole and iopamidole were detected at µg/L range (Figure 3.7), the later varies largely between < 1 µg/l – 5 µg/L depending on the location. No clear correlation was identified between the content of ICM's and the bulk parameter absorbable organic iodine values (AOI), which varied between 10 and 30 µg/L in abstracted groundwaters.
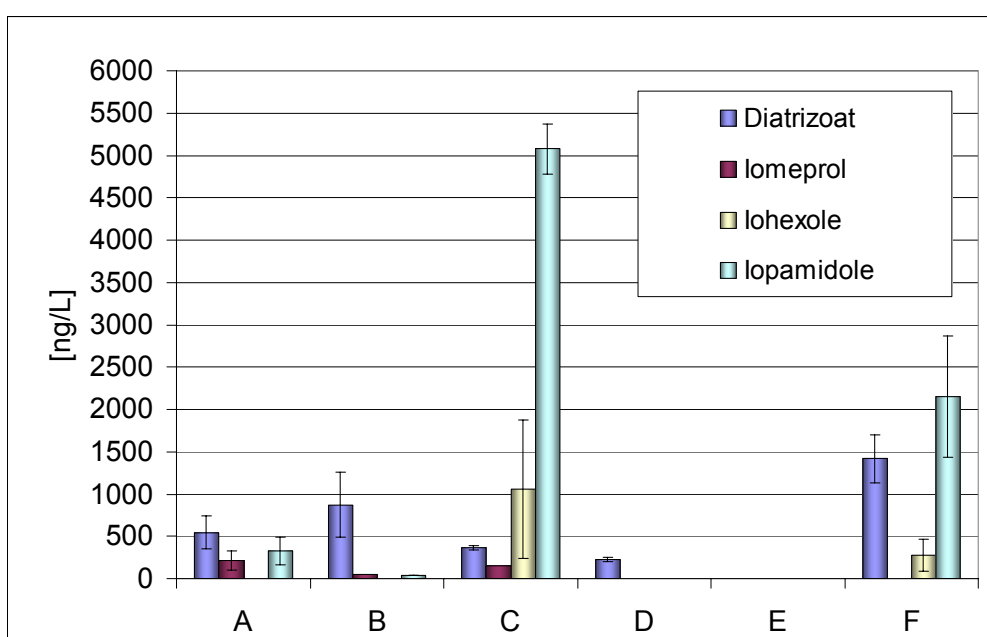

Figure 3.7 Product well ICM concentration at six SAT sites

For investigated sulfonamide and macrolide antibiotics (sulfamethoxazole, sulfamethazin, clarithromycin, roxithromycin, erythromycin and trimethopim) concentrations in effluents were found to vary from 50 – 1300 ng/L, depending on the degree of treatment. In the abstracted groundwater much lower concentration were detected, depending on the SAT conditions (especially defined redox conditions are important for effective removal) as well as the retention time, ranging for single compounds from 0 – 300 ng/L.

A set of nine different N-nitrosamines in effluents have been detected where disinfection is applied; concentrations varied between 0.5 - 55 ng/L, with higher fluctuations in the different measurement campaigns. Results identified optimisation potential for disinfection practice; the results were directly linked with investigations within WP5 performed by EAWAG. In groundwater only two nitrosamines compounds were found at one site at a very low concentration. Moreover other trace organic compounds have been measured and detected mainly in effluents and first treatment steps such as bisphenol A, estrone, ibuprofen,

diclofenac, naproxen, benzafibrate, benzotriazole and tolyltriazoles. Details can be taken from the final milestone 3.2b.

In parallel to the single compound analysis, WP3 characterized the different sites and samples by organic matter measurements as bulk parameter and its further characterisation by size exclusion chromatography (SEC) with organic carbon, UV and total nitrogen detection. Moreover fluorescence measurements (excitation emission measurements, EEM) were conducted. The content of bulk organic carbon (DOC) in the secondary effluents ranged from 5 - 15 mg/L, and in the final ground waters from <1 – 5 mg/L. Average DOC removal rates for secondary effluent to final product water are in the series A-F: 51%; 65%; 75%, 90%, 75%, 15% (Figure 3.8).
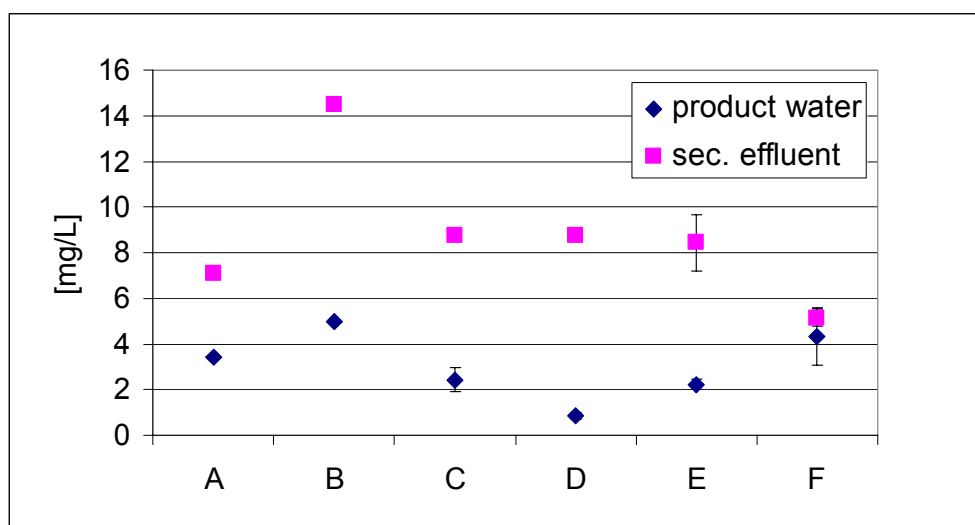

Figure 3.8 DOC of effluent and final groundwater at six different demonstration sites for groundwater recharge
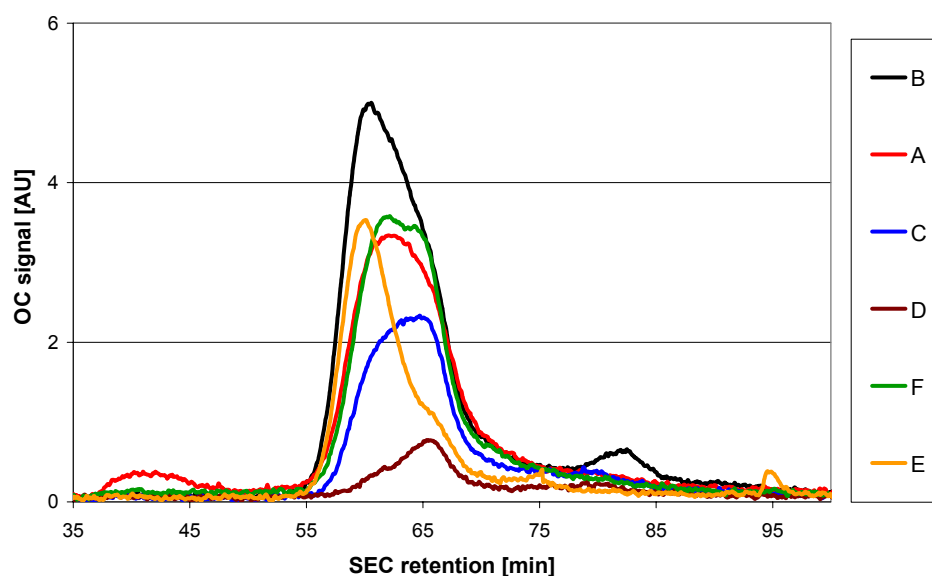

Figure 3.9 DOC characterisation by size exclusion chromatography in abstracted groundwater at six different SAT sites

The succession of different fractions of the DOC can be seen in Figure 3.9, in which content of DOC vs. molecular weight in the final product water is plotted. Even if bulk DOC values are comparable, the distribution of organic compounds may vary largely.

Due to matrix effects and total resolution, the specific identification of single organic compounds is not possible by SEC. Nevertheless organic groups and their distributions in product waters can be identified. Next to organic carbon detection the parallel detection of UV254 and total organic nitrogen offers further insights and allows tracking of unsatured organic constitutent as well as of protein like compounds.

In summary, SEC and EEM offer the potential to identify specific fractions originating from wastewater or natural water (effluent organic matter vs. natural organic matter) and can describe their fate and behaviour during water treatment. Besides single compounds that could be identified as wastewater tracers such as carbamazepine and diatrizoate the analytical combination of SEC and EEM provides evidence of specific organic fractions originating from domestic wastewaters.

WP4: Technical water reclamation and aquifer recharge case studies

METHODOLOGY

For this project, the different sites (described in the introduction, cf. Table 3.1) have been periodically monitored over two years except for the Mexican and South African sites, which entered the project later and thus have been monitored over one year only. The monitoring covered basic wastewater parameters, microbiological parameters, trace elements and salinity related parameters (Table 3.3) that were analysed in secondary or tertiary treated effluent, prior to injection or soil aquifer treatment (SAT) application and in the abstracted water (Table 3.4).

Table 3.3   Parameters analysed at the case study sites ('protocol 1')

Basic analysis	Microbiological analysis	Trace elements analysis	Salinity related analysis
Suspended solids, BOD, COD, DOC, Ammonia, Nitrite, Nitrate, Total N, Phosphorus, Alkalinity, pH, Turbidity, Detergents, Mineral oils, Phenols	Total bacteria count, Faecal coliforms, *E. coli*, Enterococci, *Clostridium* spores, Bacteriophages	Boron (inorganic tracer), Cadmium, Chromium, Cobalt, Copper, Fluoride, Iron, Lead, Manganese, Molybdenum, Nickel, Selenium, Barium, Cyanide, Zinc, Hexavalent chromium and sulfide	Chloride, Electrical conductivity, Sodium, Potassium, Calcium, Magnesium, Carbonate, Bicarbonate, Sulphate

Table 3.4 **Sampling point description**

	Case study	Source	Injectant	Abstracted water
	ITALY	Secondary effluent	Secondary effluent after transportation via open channel	Water from well located 500m away from injection point
	SPAIN	Secondary effleunt	River water (mixed with secondary effluent)	Water recovered in a mine under the river bed
ISRAEL	**CONV. SAT**	Secondary effluent	Secondary effluent	Water abstracted via wells located 300-1500m from the recharge basins
	UF PILOT SITE	Secondary effluent	UF effluent	Water abstracted after rapid SAT in wells located 7.5 and 17.5m away from injection
	BELGIUM	Secondary effluent	UF+RO treated effluent in infiltration pond	Water abstracted via wells after dune filtration
	CHINA	Secondary effluent	Ozonated and sand filtrated effluent	Water monitoring via wells
	AUSTRALIA	stormwater	Wetland treated stormwater	Water recovered from set of 4 wells situated 50 m away from injection wells
	SOUTH AFRICA	Secondary effleunt + stormwater	Secondary effluent + stormwater in infiltration basins	Water abstracted via wells
	MEXICO*	Raw wastewater	Raw wastewater	Water recovered from wells, or dug wells, or forming springs

*The results presented in this section are for the area where irrigation has been practised for 50 years

RESULTS

Over the duration of the project, the data collected at each test site have been gathered in a common spreadsheet and analysed by determining, for each parameter data set, the average value, the minimum and maximum levels observed, and when applicable the median, the lower and upper quartile values.

The unique character of this study resides in the variety of the sites involved in term of sources used for recharge, pre-treatment options, aquifer recharge methods, aquifer types and reuse purposes. Comparing the sites in term of their performances in recharging aquifers with reclaimed effluent for reuse is challenging as expectedly a multitude of factors are susceptible to influence their efficiencies (Figure 3.10).

Indeed, as highlighted by Dillon et al. (2008) in their critical evaluation of combined engineered and aquifer treatment systems in water recycling, the source of reclaimed water to be used for recharge, hence its level of pre-treatment, will depend on a combination of several factors which include the hydrogeology of the aquifer, confinement of the aquifer and the quality of the native groundwater. In his report on "criteria for health related guidelines" for aquifer recharge, Brissaud, (2003) also suggested that the quality of the reclaimed water to be used for recharge should be adapted to the characteristics of the site (i.e. recharge method, hydraulic load, infiltration schedule) and the intended use of the abstracted water post recharge. For example, for indirect potable supply, the quality of the injectant should meet requirements for drinking water supply, while for an aquifer used for irrigation purposes the quality of the injectant should meet the standards for irrigation. In this context, aquifer recharge should not be viewed as a treatment step, but an additional barrier to treatment.
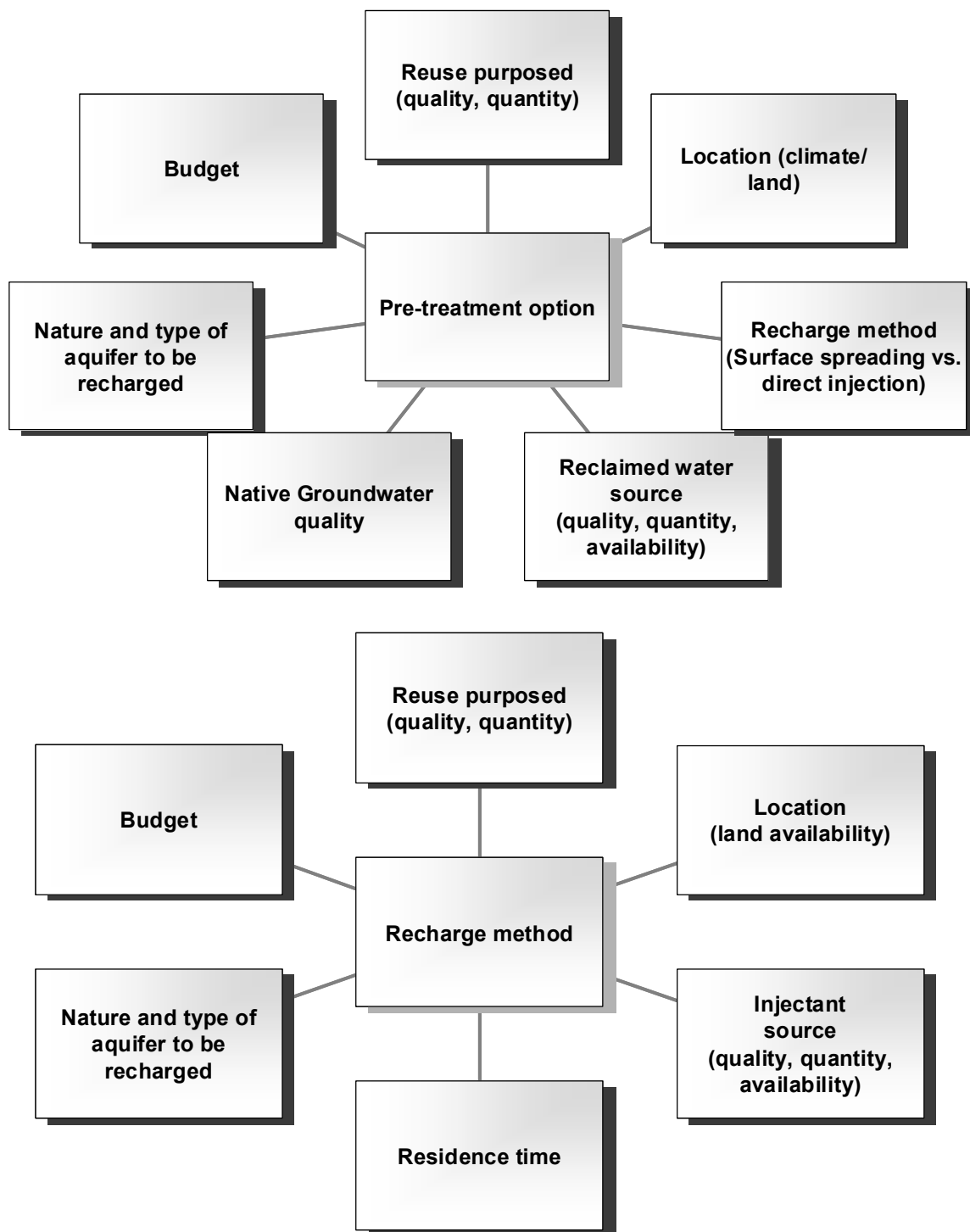
Figure 3.10 ***Factors affecting pre-treatment and recharge method choices for aquifer recharge of reclaimed effluent.***

Wulpen/Torreele, Belgium

In Belgium, the reuse of tertiary treated municipal wastewater, combined to groundwater recharge, resulted in a sustainable groundwater management of the Flemish dunes. Indeed, at this site the combination of UF and RO membranes guaranteed an excellent quality of the infiltration water and confirmed that a multiple barrier approach, which means different techniques are combined, assures a safe solution when drinking-water is the targeted reuse purpose of aquifer recharge schemes.

Nardó, Italy

In Italy, the monitored aquifer recharge scheme consisted in the sinkhole infiltration of secondary treated effluent in a karstic aquifer. Overall, this study showed that the quality of the water recovered in wells can be considered as a function of the aquifer transmissivity and particularly of the injectant water quality. If injectant quality is inferior to the existent groundwater quality, the reclamation of water may lead to the accumulation of pollutants (such as nitrates, THMs, organic compounds) and salts that will produce unusable ground water at recovery wells. The experimental studies suggest an integration of the actual water treatments including a new treatment plant before the injection into the sinkhole. The treatment can be selected by considering appropriate technologies and by taking in consideration the efficacy of the soil aquifer treatment of the Nardò subsoil (i.e. disinfection and biodegradations capacity). Overall, this site proved efficient as a barrier against salt water intrusion.

Sabadell, Spain

At the Spanish site, where secondary treated effluent are used for aquifer recharge via river bed filtration and reuse for irrigation of a public park and street cleansing, the study showed that the artificial recharge system is able to reduce substantially the basic wastewater parameters (DOC, COD, Suspended solids), macronutrients, and all the microbiological parameters by 3 to 4 log units. Salinity analysis also showed that the water recovered had very high proportion of water infiltrated from the river basin. Overall, the use of treated wastewater for aquifer recharge at this site helped preventing water restrictions and provided an additional source of water for street cleaning. Although the abstracted water is not used for agricultural reuse, the relatively high salinity levels observed along the scheme could be a source of concerns as it could affect the soil properties at long term.

Shafdan, Greater Tel Aviv, Israel

In Israel, a pilot site using ultra-filtrated secondary effluent from the Shafdan WWTP for short Soil Aquifer treatment (SAT) was compared to the performance of a 30 years old conventional SAT system. At this site, the aim was to look at alternative technologies using high effluent quality for infiltration, short retention time systems with low footprint that would produce reclaimed water for unrestricted irrigation qualitatively comparable to the accidental drinking water quality of the long term conventional SAT abstracted water. The analysis of the final results observed at the pilot site showed that compared to removals obtained at the conventional SAT, micro-organisms removals were similar, organics removals (as total DOC) were efficient although slightly less effective than at the conventional SAT, and nitrogen levels almost similar. However, issues regarding redox conditions remained (*i.e.* manganese dissolution) and solutions such as producing conditions for minimum bacterial activity and maximum oxygen may help alleviating this issue.

Salisbury, Adelaide, Australia

At the Australian site where wetland treated stormwater was used for aquifer storage transfer and recovery (ASTR) in a brackish aquifer, the monitoring of the site showed that the water sampled at the outlet of the reed bed system can meet drinking water standards except for iron, colour and microbiological indicators. In 2009, stored stormwater was abstracted from the ASTR and preliminary results indicated that with the exception of colour, turbidity, iron and manganese, Australian Drinking Water Guidelines (2004) could be met. This study confirmed the great potential of ASTR systems in producing high quality water for non-potable reuse purposes. However, the role of ASTR in producing constant reliable supplies of drinking water will require further investigations including quantitative risk assessments and further monitoring after several cycles of injection and recovery.

Gaobeidian, Beijing, China

In China, the operation and monitoring of the pilot site where advanced treatment of municipal effluent via coagulation, ozonation and sand filtration was used for direct aquifer recharge, showed that ozonation contributed to biodegradability improvements while the slow sand infiltration further removed organics (as DOC). The combination of slow sand infiltration and well recharge proved sustainable and would be particularly suitable for areas with low surface infiltration rate. Overall, the compact character of this site (*i.e.* low footprint) ensuring the treatment of surface recharge for future reuse is promising for a megalopolis like Beijing which suffers from severe water shortage as a consequence of the increasing population but where land availability is low.

Tula Valley (El Mezqital), Mexico City

At the Mexican site, unintentional aquifer recharge is the result of the long-term irrigation (*i.e.* 10- 100 years) of agricultural land with raw wastewater. The wastewater entering the Tula Valley from Mexico City contains high concentrations of a variety of contaminants including microorganisms, organic micropollutants, salts, and various metals. The atypical natural soil aquifer treatment removes a vast majority of most contaminants, and results in a source of water that complies with most potable water criteria. However, the monitoring of the sites showed that some concerns remain regarding the quality of the water as for example most of the microorganisms evaluated were detected infrequently in wells, springs, and dugwells; the supply water is at least as saline as the wastewater; nitrates, total dissolved solids, fluorine and aluminium exceeded regulatory limits in some sources of water while metal accumulation in the soils is occurring although regulatory limits have not yet been exceeded. The study then suggested that an advanced treatment such as nano-filtration would be necessary to generate a trustworthy potable water supply.

Atlantis, Cape Town, South Africa

In South Africa, the results showed that indirect recycling of stormwater and treated domestic wastewater via pond infiltration augments the limited groundwater supplies in a publicly acceptable manner but water quality management remains the dominant issue regarding water supply at Atlantis. Overall, the Atlantis system seems to be quite robust from a water quality point of view. However, the presence of iron in the groundwater is of concern due to the clogging of the production boreholes. From a microbiological point of view the subsurface passage plays a decisive role as a safety barrier. Overall, the Atlantis groundwater scheme provides a cost-effective water supply option when coupled with strict management of the resource. The importation of limited quantities of low salinity surface water has enhanced the viability of the recharge scheme and also allows the utilisation of slightly more saline groundwater. The future introduction of membrane processes for water softening and partial

desalination will further enhance the scheme, even allowing the possible export of potable water to other residential areas.

CROSS CASE ANALYSIS

Despite an inevitable case by case approach when planning aquifer recharge as widely explained in the literature (Dillon and Jimenez, 2008; Bouwer, 1996, Franson 1988), the recharge and reuse practises studied here present technical aspects similar to a large number of schemes in the world. The performances of the sites tested and the difficulties encountered during the study should then provide valuable information for organisations planning or operating MAR sites.

When comparing the site in terms of injectant source, the sites either used raw wastewater as injectant at the unintentional soil aquifer treatment system in Mexico; secondary effluent as injectant in Italy, Spain, Israel (i.e. conventional site) and South Africa (blended with stormwater); advanced treated effluent as injectant in Belgium (RO+UF), Israel (UF), China (Coagulation-Ozonation-Sand Filtration) or wetland treated stormwater in Australia.

As described above, the level and type of water reclamation technologies applied prior to recharge unsurprisingly determined the contaminant load needing polishing by the aquifer. However, the aquifers preceded by lower levels of pre-treatment (i.e. secondary effluent, or raw wastewater) seem to be able to buffer high contaminant concentrations and achieved an abstracted water quality comparable to sites using high pre-treatment technologies. Though, when analysing the actual impact of pre-treatment level on basic parameters removals rates after recharge (Figure 3.11), as expected removal rates after passage through soil typically decrease, when the level of pre-treatment increases. This corroborates with the fact that independently from the recharge method and the type of aquifer used, the aquifer primarily acts as a treatment step when lower quality of effluent are used, while it acts as a polishing step when higher effluent quality is used.
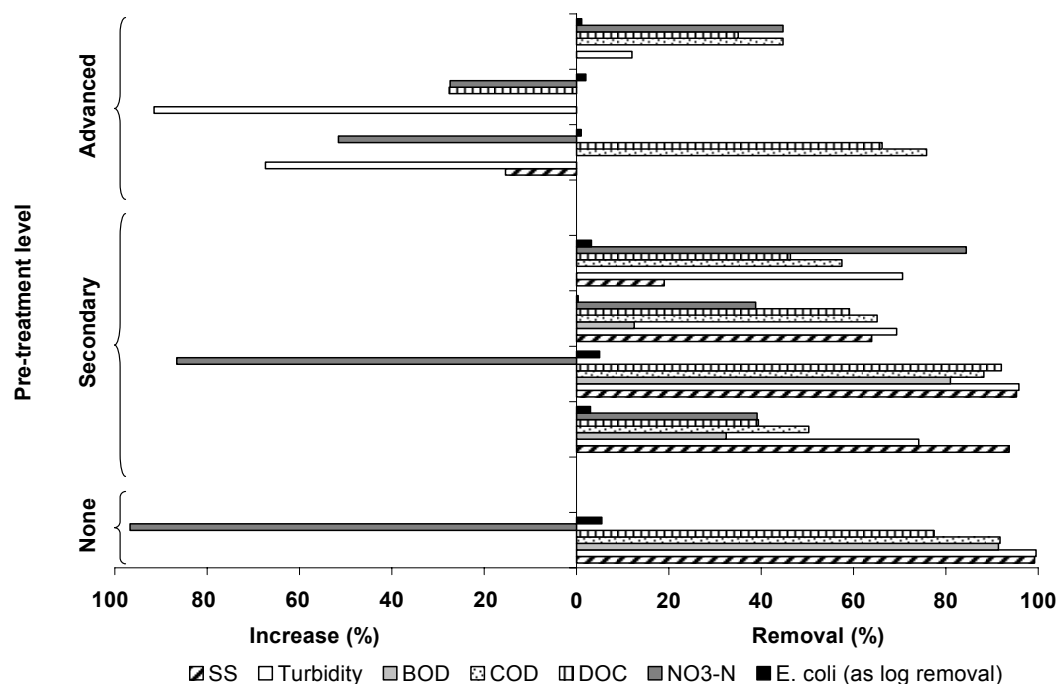

Figure 3.11 *Influence of the injectant pre-treatment level on average percentage removals or increase after aquifer recharge at each site.*

The residence time that the injectant spends within the aquifer before recovery could also be one of the factors enhancing the capacity of the sites using secondary effluent to achieve removals comparable to sites using advanced technologies. For example, the Israeli conventional SAT site uses secondary effluent that travels within the aquifer for a minimum of 180 days to a maximum of 360 days, hence providing higher opportunities for removal than at the pilot Israeli site where highly treated effluent can lead to higher infiltration rates in the aquifer (Chilton and Alley, 2006), hence also reducing aquifer travel times and chances for quality improvements. Similarly, in Belgium the level of pre-treatment prior infiltration ensuring robust, reliable treatment of effluent to high quality standards prior recharge combined with the 35 days retention time leaves lower opportunities for quality improvements during infiltration. Overall, when comparing the performances of the site in term of removal of basic wastewater parameters versus retention time in the aquifer (when available) independently of the recharge method used, no particular trend could be confirmed (Figure 3.11). Another important fact in the cross case comparison is that in those MAR schemes being injected with advanced treated water e.g. RO permeate, the concentrations of certain standard parameters can easily increase. This remineralization is intentional, in particular when the water is used for drinking water production.

When comparing the influence of the residence time (Figure 3.12) the removal performance seemed lower for retention times below 60 days.
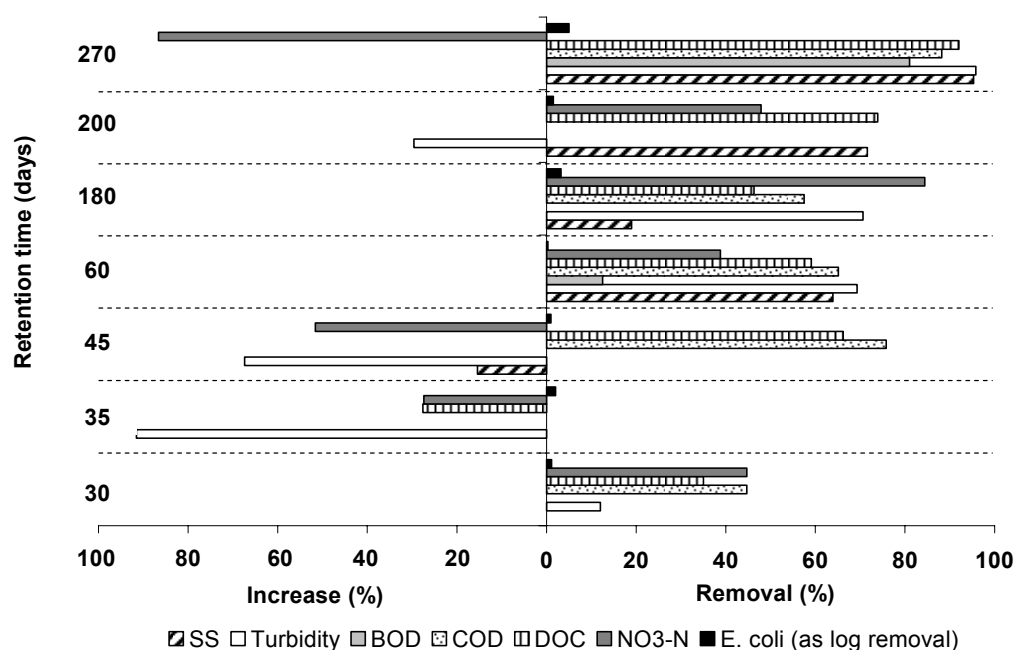
Figure 3.12 *Average percentage removals or increase associated to the retention times of the sites studied.*

The particular case of the Australian site where wetland treated stormwater is used for recharge by means of ASR makes it difficult to compare to the other sites. Indeed, stormwater usually present lower contents in dissolved solids, nutrients, oxygen demand and salinity and higher SS, heavy metals and bacteria level when compared to wastewaters (Barnett et al., 2000). However, this study showed the good capacity of wetland treatment to act as a reliable and robust polishing option for upgrading stormwater for groundwater recharge, hence

helping to reduce the salinity of brackish groundwater and increasing options for reuse. Indeed, monthly injection rates of 16400 m³ generally met Australian Drinking Water Guidelines (2004) except for turbidity, E. coli, Enterococci, faecal coliforms, and iron, while after conditioning of the aquifer the stored stormwater met drinking standards (exception for total iron).

CONCLUSIONS

No major operational issues were observed over the duration of the project although the study showed that some problems remain regarding the control of redox conditions within the aquifer which can cause maintenance and clogging problems as illustrated in Israel and Spain. However, this study confirms the benefits of using reclaimed wastewater or stormwater for aquifer recharge with the ultimate objectives of combating water restrictions (Spain), preventing salinity and seawater intrusion (Italy, Australia, South Africa), and producing high water quality for irrigation (Israel) and potable reuse purposes (Belgium, South Africa).

The project outputs did not specifically allow drawing out some definitive conclusions on the type of pre-treatment required for a specific type of aquifer recharge site and reuse purpose or on standard residence times that should be respected for specific types of infiltrated water quality. For example, suggesting that the performances obtained through the infiltration of secondary effluent via soil aquifer treatment under specific operation conditions will be reproducible at another site operated under similar conditions is not feasible. Similarly, adopting a conservative approach such as treating influent used for recharge to a high standard may not be adapted to other type of sites. It confirms then that a global approach for the selection of pre-treatment options and recharge methods for managed aquifer recharge to ensure their reliability, robustness and sustainability in treating and storing reclaimed water for a determined reuse purpose is not recommended.

Deriving reliable policy advice and recommendations from the knowledge generated through these case study sites is therefore very problematic. Aquifer recharge and reuse schemes are highly complex undertakings involving multi-stage treatment processes utilizing a variety of physcial, biological, and chemical interactions. Operational and climatic conditions strongly influence scheme performance and our suite of case study sites involved a variety of source waters and reuse applications. Consequently, an evidence based assessment which allows scheme planners and designers to select 'Configuration X' or 'Treatment train Y' (even for a specific resue application) is not possible. Even qualitative judgements about 'better / worse' or 'more / less appropriate' technologies or techniques for specific applications are irrelevant given the different ambitions of the monitored schemes and their operational variation.

This is not to say that we are unable to draw any conclusions. The detail of local climatic and operational circumstances coupled with uncertainties associated with aquifer dynamics and performance force an iterative approach to the design and implementation of recharge / reuse schemes. The case studies illustrated above have all recognised this and have taken appropriately conservative approaches to scheme development. We might thereby characterise 'appropriate' schemes as pursuing structured exploration of the recharge / reuse system dynamics, focusing on issues of reliability and risk. Yes, information and experiences from other, similar schemes may highlight particular areas of concern. But the solutions applied in similar schemes are unlikely to be wholly relevant. Success can then be stated in terms of fitness for purpose rather than conformity with a global standard. Interestingly, this raises a secondary challenge of being able to adequately evaluate whether the purpose (in terms of intended outcomes) of a recharge / reuse scheme is achievable prior to a major investment being made. The iterative approach suggested above allows such an evaluation

during pilot trials etc. but the management model here is not standard. Decisions on scheme ambitions and performance need to be informed by data generation activities which may constrain those ambitions. All this suggests that costs may be high before a decision can be reached on scheme feasibility … let alone scheme design and operation.

WP5: Specific studies on removal and fate of microorganisms and organic compounds

METHODS AND METHODOLOGY

Soil batch experiments

The sorption experiments were performed in accordance with the OECD Test Guideline 106. Air-dried sediments with particle sizes < 2 mm obtained by sieving were used in batch experiments. Well known European standard soils such as Lufa 2.2 and Euro Soil 5 were used as well as dune aquifer sand collected in wastewater reclamation area for groundwater recharge at the Belgium North Sea coast. Sorbents were hydrated in test solutions containing either 0.01 M $CaCl_2$ or effluent of the WWTP respectively. The biological transformation of the test compounds was carried out for aerobic conditions according to OECD Guideline 307 and to the Zahn-Wellens-Test after DIN EN ISO 7827 (corresponding to the die-away-test after OECD 301 A) and for anaerobic conditions according to Shelton and Tiedje (1984). The sorption properties of antibiotics were determined as well as of wastewater indicator substances such as the antiepileptics (carbamazepine, primidone) and the corrosion inhibitor benzotriazole.

Soil column experiments

Miscible-displacement experiments conducted in sand columns was carried out to obtain necessary information concerning the fate and mobility of several pharmaceutical compounds during subsurface passage. Soil columns experiments were carried out to explore the fate of antibiotics, NDMA, AOI as well as organic wastewater indicator substances. For the column setup industrial sand as well as aquifer material from the Wulpen site in Belgium has been selected, because it has optimal characteristics i.e. high sand content and low content of clay and organic matter.

Chemical Analysis

Trace analytical methods including accelerated solvent extraction, solid phase extraction (SPE) followed by LC tandem MS were used to determine the analytes in water as well in the soil matrices (Schlüsener et al., in preparation; Krauss and Hollender, 2008).

Nitrosamine formation. The potential to form nitrosamines was studied for whole wastewater samples and physico-chemical wastewater fractions of a case study site. The fractionation protocol consisted of sequential filtration steps to <1 μm, <0.2 μm, and <2,500 Da, followed by a fractionation over C18-modified silica gel to separate a hydrophilic (permeate) from a hydrophobic (retentate) fraction. The nitrosamine formation potential (NA-FP) as a proxy for the sum of all nitrosamine precursors in bulk samples and fractions was determined as the nitrosamine concentrations formed after excessive chloramination.

RESULTS AND CONCLUSIONS

Soil batch experiments

Affinities of all analytes to the soil increased from Wulpen sand, over Lufa 2.2 to Euro Soil 5, indicating that the organic carbon contents might be crucial for sorption. Isotherms were well described by the Freundlich model. Sorption was mainly linear or close to be linear (n=0.93-1.07) for most target compounds and soils. All sorption studies confirmed that the selected organic substances are appropriate to be used as wastewater indicator substances with respect to their sorption behaviour. In water/soil systems, benzotriazole and primidone as well as the iodinated contrast media (ICM) iomeprol, iopamidol, iohexol and iopromide were bio-transformed into several transformation products (TPs), while the ICM diatrizoate, carbamazepine were stable over more than 100 d. For iopromide 12 TPs could be identified. *Therefore, only diatrizoate and, carbamazepine can be recommended as wastewater indicators, while sulfamethoxazole, benzotriazole and primidone could be measured in parallel but the results have to be taken with cautious.*

Soil column experiments

A breakthrough of NDMA and Sulfamethoxazole (SMX) was observed through the columns of sand from the Wulpen site. For NDMA the break through reaches about 60 % indicating that NDMA is being degraded in the soil column. The SMX effluent concentration reached 100 % and seems not to be degraded. The degradation of NDMA was expected, based on our batch studies applying 14C-labeled NDMA. NDMA appears initially to be degraded during passage, however, the concentration of NDMA increases again indication either a lower degradability or an interaction with SMX which was added later to the soil column. No major differences for both redox and temperature controlled columns could be observed with respect to the removal of sulfonamide and macrolide antibiotics. All macrolides were nearly completely eliminated (> 80%) in all columns independent of the temperature and the redox conditions. Sulfonamides are more incompletely removed. Sulfamethoxazole and trimethoprim were in general up to a level of 30% better removed than sulfamethazine with up to about 55%. The breakthrough curves of carbamazepine, primidone, sulfamethoxazole, and benzotriazole confirmed that sorption can be neglected, but that in some cases such as iopromide biodegradation has to be taken into account as found in the batch experiments. Soil column experiments have also been performed to assess the fate of bulk effluent organic matter (EfOM) in soil column experiments simulating soil passage. Results derived from fluorescence excitation emission matrix (F-EEM) and liquid chromatography with organic carbon detection (LC-OCD) indicate preferential removals of protein-like and polysaccharide-like organic matter over humic-like organic matter. This result has implications for post-treatment by membrane filtration (fouling reduction) and chlorination (disinfection by-products), and as a carbon source(s) for driving heterotrophic biodegradation of organic micropollutants.

Nitrosamine formation

The formation potential for N-nitrosodimethylamine (NDMA-FP) was by far the largest of all nitrosamines studied and as high as 4500 ng/L in STP influent, and 900 ng/L in STP effluent. About 70% of the NDMA-FP in influent samples could be attributed to rather polar low-molecular weight precursor compounds (e.g., dimethylamine). Of these compounds, about 90% were degraded during activated sludge treatment. In contrast, more hydrophobic precursors were degraded to about 75% during secondary treatment. Higher-molecular weight compounds like polymers or humic substances contributed to less than 15% of the NMDA-FP in the effluent and were removed during the ultrafiltration step. The reverse osmosis was very effective and rejected more than 98% of the NDMA precursors. The formation potentials of the other nitrosamines decreased to the same extent during treatment than those of NDMA. The results indicate that it is unlikely that nitrosamines are formed if reclaimed wastewater is

chlorinated or chloraminated during drinking water production if sophisticated purification steps such as reverse osmosis are used.

Ozonation experiments

Most of the selected compounds were very efficiently oxidized via ozonation. However, the identification of the oxidation products formed is still a challenge but first suggestion for the identification of their chemical structures can be made. Ozonation with chlorinated and non-chlorinated WWTP effluent showed an increase of ozone stability but a decrease of hydroxyl radical exposure in the sample after chlorination. This may shift the oxidation processes towards direct ozone reactions and favors the degradation of compounds with high rate constants kO3.

WP6: Case-study related risk studies

There is a complex relationship between how reuse is carried out and the associated risk. It is necessary to know what the risk is and what risks society is prepared to accept. This is followed by risk management and risk communication. The degree of risk is often inversely associated with the cost of the wastewater treatment.

Risk assessment is performed using a quantitative approach; in most of the case studies, the approach taken has been probabilistic (stochastic) at least for the pathogenic hazards. Risk management has been applied using the HACCP (Hazard Analysis and Critical Control Points) framework, which is the one adopted and recommended by the World Health Organisation. The risk assessment and the risk management consider the recharge system as well as the barriers, planning procedures and analytical work.

Both risk assessment and risk management have been applied to four of the test sites of Reclaim Water project, namely Atlantis in South Africa, Salisbury in Australia, Sabadell in Spain and Torreele/Wulpen in Belgium. After studying the four sites and performing a risk assessment, the risk management has been implemented and is now being validated or is in the process of being validated.

Risk communication has been developed widely in some case studies (Adelaide, Australia) while it is under development in others (Sabadell, Spain). A risk communication tool that is applicable to any recycled system has been provided, thus it can be applied to Managed Aquifer Recharge systems.

For all the case studies, the results of the monitoring campaigns performed in WP2, WP4 and WP5 were used to evaluate the risk. According to the tasks defined in the Reclaim Water project, several protocols of analysis were established and include microbial and chemical parameters. Apart from the classical parameters (*E. coli,* pathogenic helminth eggs, electrical conductivity etc.), pharmaceuticals, oestrogen mimicking substances, antibiotic resistance genes, pharmaceutical compounds, etc. were also assessed.

Sabadell (Spain).

Critical Control Points (CCPs) and Points of Attention (POAs) were established as one of the first steps to implement a HACCP system (Ayuso-Gabella et al., 2007). A summary of them appears in Table 3.

A deterministic quantitative microbial and chemical risk assessment was performed, following which, it was deemed necessary to perform a probabilistic risk assessment for the pathogenic hazard. For this purpose, @RISK software was used and Monte Carlo simulations

were performed. In Sabadell, exposure is considered through accidental aerosols inhalation via irrigation of lawns and gardens in parks and street cleaning.

Two approaches were undergone, both of them considering the pathogens recommended in the WHO Guidelines for Wastewater Reuse (2004) and the Australian Guidelines for Water Recycling (2006) (*Cryptosporidium*, *Campylobacter* and Rotavirus):

1. The log removals for every barrier were described by a distribution function using data from the literature. To model the SAT barrier, the log removals were described as the sum of 2 treatments:

   - The retention by the river bed was assimilated as a sand filtration treatment.

   - The residence time in the aquifer and the related decay suffered by the pathogens there was the other one.

2. The log removals were described as a Triangular distribution function with minimum, maximum and most likely values which were inferred from the indicators data generated in the Reclaim Water project. The indicators used for every pathogen were:

   - *Escherichia coli* as a surrogate for *Campylobacter*

   - *Clostridium* spores as a surrogate for *Cryptosporidium*

   - Somatic bacteriophages as a surrogate for Rotavirus

Approach 1 gives as a result a very low level of risk in the system, much below the recommended DALYs (Disability Adjusted Life Years) value given in the guidelines. In contrast, approach 2, in which log removals from indicators are used, comes out with risk higher than the recommended for all the pathogens. Both approaches are robust in terms of the results, as the mean and the 95th percentile of the probability distribution results for a selected pathogen are in the same range. However, approach 2 uses real data to describe the performance of the different treatments in the system, so this approach is giving more reliable results.

On the side of the chemical compounds, salinity was identified of a major concern in the system. The recovered water is used for irrigation of an urban park, and, in the long term, the soil and the plants can suffer from this hazard. Until now, this has not been an issue.

There has been a lot of controversy regarding the recommended value of DALYs for the different uses of recycled water, especially regarding water reuse for irrigation; some groups consider that 10^{-6} DALYs is a too restrictive value. However, it might not be enough to protect immunocompromised populations. The irrigated urban park in Sabadell is located behind a hospital, and the case of the immunodepressed or immunosupressed people is remarkably important there. More research is required in this area, and by now, ensuring a good performance of the system and a proper disinfection are the steps to take.

Finally, risk characterization in Sabadell requires a better knowledge of the hydrogeology in the area and the dilution factor in the river. Following efforts in improving the management of Sabadell sytem should be directed to these two topics.

Preventive, operational and corrective measures are being implemented according to the results of the risk assessment and CCPs identified in the system (see Table 3).

Table 3.5   CCPs in Sabadell, Spain

CCP/POA	Description	Corrective measures	Preventive measures
CCP1	Disinfection system	1. Stop pumping system 2. Check UV lamp functioning 3. Review meteorology records during previous days (e.g. rain) 4. Review any other potentially harmful events occurred in the river or the effluent of the WWTP during previous days (e.g. collector breakage, malfunction of WWTP, etc.) 5. Repair any possible breakage and perform an additional sampling to assure that the problem has been solved	Cleansing of UV lamps – monthly Chlorination probes testing – bimonthly (when the residual chlorine is measured) Microbiological monitoring of samples after UV and after chlorination for the monitoring schedule)
CCP2	Storage tank	1. Stop pumping water from the storage tank 2. Additional cleansing of the tank	Cleansing of the tank twice per year Microbiological monitoring of samples
CCP3	Sprinklers	1. Stop pumping water from the storage tank 2. Repair the sprinklers	Checking of proper function of the sprinklers – weekly
CCP4	Soil of the park	1. Stop irrigation of the park with recovered and disinfected water. 2. Check salinity of the water and decide whether to mix tap water and recovered water	Monitoring of the soil of the park
CCP5	Plants of the park	1. Stop irrigation of the park with recovered and disinfected water. 2. Check salinity of the water and decide whether to mix tap water and recovered water	Monitoring of the plants of the park
POA1	Ripoll River WWTP effluent	1. Discharge of the water downstream of the infiltration area 2. Check every step of the process, especially the secondary treatment (e.g. sludge losses)	Assure a proper functioning of the WWTP Upgrading the applied treatments
POA2	Ripoll River	1. Improve the effluent of the Ripoll River WWTP by a tertiary treatment (will be done in the near future) 2. Develop a general plan jointly with the municipalities upstream the river to prevent illegal discharges and to improve the quality of the WWTP effluents discharged	Monitoring of the river in different points
POA3	Groundwater	1. Stop sending the Ripoll River WWTP effluent to the river 2. Check illegal disposals	Monitoring of groundwater in different points
POA4	Distribution system	1. Stop distribution of recovered and disinfected water 2. Purge the system	Assure a good disinfection of the water and a low level of organic matter

Atlantis, Cape Town (South Africa).

A deterministic quantitative microbial risk assessment was performed, following which, it was deemed necessary to perform a probabilistic risk assessment. For this purpose, Analytica software is being used. Monte Carlo simulations have been performed. In Atlantis, to predict human health risks through direct consumption of the recharged treated wastewater, treatment efficiencies are used, calculated with real data as well as with data from the literature.

Three distinct approaches were applied to quantify microbial risks:

1. Firstly, positive counts of *Cryptosporidium* and *Giardia* from the monitoring program were used to describe the concentration of these pathogens in treated wastewater effluent. The recharge water treatment train was assumed capable of achieving a 3 log reduction as required in the US EPA's Drinking Water Regulations.

2. A second approach was used, given the low number of detects for Cryptosporidium and *Giardia*. Sulphite reducing clostridia (SRC) counts were used as an index for *Cryptosporidium*. Results from the Atlantis system showed the concentration of *Cryptosporidium* in the treated wastewater effluent at a ratio of *Cryptosporidium*: SRC equal to 1: 106. The two approaches mentioned above made use of the exponential model for calculating the risk of infection to parasites.

3. Thirdly, an epidemiologic approach was applied to estimate *Cryptosporidium*, *Giardia*, *Salmonella* and Rotavirus concentration in sewage, based on the incidence of infection in the community, duration and intensity of pathogen excretion (Ashbolt et al., 2006; Gerba, 2000).

To quantify the chemical risks, endocrine disruptors in the form of oestrogen mimicking substances were assessed, as well as pharmaceutical substances.

Results for the microbial and chemical quantitative risk assessment point to the importance of the subsurface passage as a safety barrier in the system. The key barrier was unsurprisingly the aquifer, illustrating that the integrity of the infiltration basins must be maintained and that abstraction rates of groundwater must be carefully controlled. Stormwater is also a contributor to possible risks and must also be incorporated into the HACCP plan. The QMRA was a useful tool in the management of the Atlantis artificial recharge scheme and was used in the compilation of a HACCP process described in the next section for the management of these risks. The same was found for the chemical analyses of pharmaceutical compounds and oestrogen mimicking substances. The following points were identified as important drivers of risk via the risk assessment:

- If the various barriers and in particular the infiltration basin, are not in place a high probability of infection from parasites may take place. This probability of infection may be as high as 1 in 1000.

- Assuring a 3 log reduction takes place in the treatment process these risks can be reduced to 1 in a 1,000,000. The retention time within the aquifer is particularly important in reducing pathogenic organisms. This was originally considered for the management of salinity.

- Stormwater (SA3) tended to have higher microbial counts than the treated wastewater effluent. This has implications for the management of the water scheme. Stormwater management to ensure integrity of the thin unsaturated zone confining the reduction capacity of the soil aquifer system during basin recharge will be important in the overall management of the artificial recharge system.

Table 3.6   CCPs, preventive and corrective measures in Atlantis

Points	Description	Hazard	Preventive action	Corrective action
CCP1	Maturation Pond inlet to monitor quality of Domestic waste water effluent entering the Atlantis Recharge System	Illegal toxic discharge	Online EC monitoring	Redirect waste water effluent before maturation ponds to industrial ponds
CCP2	Stormwater (urban runoff) from residential area	Illegal toxic discharge/ accidental spill	Online EC monitoring Trace metal monitoring (quarterly)	Redirect first flush / Redirect stormwater to industrial ponds before entering source water stream
CCP7	Final Product (Drinking Water)	Bacterial regrowth	Residual chlorine	Hold / retain water, alternative Melkbos supply
CP1	Base / low flow and peak flow diversion to recharge basins	Integrity of soil, and retention time	On-line flow meter to monitor base and peak flows	Avert between recharge basins
QCP1	Water treatment and softening plant brine transport pipeline to coastal recharge basins	Clogging/ breakage of pipe	On-line flow meter	Divert to storage tank and transport off-site for removal
QCP2	Urban run-off from Pond 5 that includes run-off from industrial area	Toxic / illegal spill	On-line EC monitoring	Divert to industrial pipeline and coastal basins
QCP3	Urban run-off including oil in Pond 9	Toxic / illegal spill	On-line EC monitoring	Divert to industrial pipeline and coastal basins

Salisbury, Adelaide (Australia)

While previously HACCP approaches were applied to the Aquifer Storage Transfer Recovery (ASTR) project (Rinck-Pfeifer *et al.* 2005) located at Salisbury in South Australia in a largely qualitative manner (Swierc *et al.* 2005), this work builds on the previous study by moving towards a quantitative risk assessment.

Starting from a preliminary qualitative HACCP plan (Swierc *et al.* 2005) for ASTR, the perusal of a risk-based water quality monitoring program forms the basis of a improved quantitative risk assessments in the HACCP framework. Periodic analysis and review of the HACCP plan also give a rationale for further fine tuning of the risk-based water quality monitoring program and associated research to support the implementation of the evolving quantitative HACCP plan for the ASTR project. Of primary importance is the continual review and improvement of the system by re-evaluating the CCPs and the supporting monitoring programs.

The potential microbial hazards identified were represented by the commonly used "index" pathogens, rotavirus, *Cryptosporidium parvum* and *Campylobacter* representing virus, protozoa and bacterial hazards, respectively consistent with current guidelines (NHMRC 2004; WHO 2004). For the pathogens, the yearly disease burden in terms of Disability Adjusted Life Years (DALYs) was calculated and compared to current acceptable thresholds (NRMMC/EPHC 2005). The QRA model was further developed to facilitate Monte Carlo simulation using @RISK. The simulation represents the inherent variability in the process of stormwater harvesting, wetland treatment and subsurface storage and treatment characteristics based on the modelling by Pavelic *et al.* (2004) and influence on expectant risk as well as the uncertainty in the mathematical model of the process.

The calculated risk is in the order of 10^{-3} DALYs which represents an unacceptable risk. The treatment capacity of the aquifer and the cleansing reed bed was assumed to be zero until suitable pathogen decay studies have been completed as part of validation monitoring. Given the likely decay rates for each of the index pathogens, each of them will achieve an important log removal during the subsurface storage treatment component. The travel time of injected water within the aquifer will be validated during the ASTR trial. Finally, the necessity or further additional treatments in order to make use the recovered water as a drinking water will be evaluated.

As per the chemical risks, the presence of organic compounds was a main concern in the system. A quantitative risk assessment of pesticides, herbicides and other organic compounds likely present in the source water was performed, and the results showed and acceptable level of risk for drinking water purposes.

Another hazard of concern in the system is the salinity, as the water is injected in a salty aquifer. After flushing the aquifer during the preliminary conditioning phase, the recovered water has an acceptable salinity for drinking water purposes.

Some other issues to be solved in the future are the turbidity and the iron. The turbidity is expected to be reduced when the system will work in a fully operational way, not only as a trial place. The iron will probably have to be removed by some additional treatment.

Table 3.7   CCPs, critical limits and corrective measures in Salisbury

Control Point	Description	Critical limit	Corrective measure
CCP#1	inlet to the in-stream basin	Turbidity < 100 NTU; Conductivity still to be developed subject to baseline water quality monitoring of the influent stormwater.	Water not transferred to the holding storage.
CCP#2	outlet of the cleansing reedbed	Turbidity still to be developed; Conductivity also still to be developed depending on mixing ratios in the subsurface.	Water not transferred to the injection well.
CCP#3	recovery well	Turbidity < 1 NTU; Conductivity still to be developed but with a maximum TDS of 500 mg.L^{-1} plus other parameters as specified for potable water quality in the Australian Drinking Water Guidelines (2004).	Water not transferred to the end user.
QCP#1	POA –inlet to the Parafield stormwater harvesting facility	None set, still subject to the HACCP validation program.	Investigation of water quality exceedence.
QCP#2	POA –inlet to the cleansing reedbed	None set, still subject to the HACCP validation program.	Investigation of water quality exceedence.
QCP#3	POA –injection well	As appropriate for potable water quality specified in the Australian Drinking Water Guidelines (2004) with respect to hazards not attenuated in the aquifer.	Investigation of water quality exceedence.

Torreele/Wulpen (Belgium)

A system management plan based on the Hazard Analysis and Critical Control Points (HACCP) concept was put in place during the planning phase of the project (Dewettinck *et al.*, 2001).

The Reclaim Water project was seen as a learning opportunity to validate the plan and reframe it in the context of the Water Safety Plan (WSP) approach, recently developed by the World Health Organisation (WHO, 2005). The WSP framework systematises long-established principles –including the HACCP concept-, and builds on scientific and managerial developments, from catchment to consumer. The WSP was developed in collaboration with IWVA.

The quantitative risk analysis (QRA) was performed in a deterministic way, using all the data generated during the Reclaim Water project and also data coming from the monitorin routine at Torreele/Wulpen. The QRA results can be summarized as:

- the multi-barrier system has a high level of reliability and it is effective at producing safe drinking water.

- RO is the major and ultimate single barrier against both microbial and chemical contamination of drinking water and the aquifer alike. Results so far show that RO offers complete removal of all pathogenic agents and can be considered as good practice for the removal of pharmaceutically active chemicals and endocrine disruptors.

- Recontamination of the infiltration pond from wildlife has also been identified as a health-bearing risk.

- The risk-containing hazard of pathogen regrowth in the distribution system is expected to be similar to that of drinking water production with conventional raw water sources. The reverse osmosis step produces a very low content of organic materials, while the biological treatment is designed to remove nitrogen and phosphorus nutrients to levels that should not encourage microbial regrowth nor bio-film persistence. The iron levels contained in the groundwater should be able to fix any low-frequency possible orthophosphate peaks arising from the pre-treatment scheme.

- Under good operating conditions, the low chloramine dosing rate used for biofouling control in combination with the low TOC levels do not seem worrying for the formation of hazardous levels of disinfection by-products.

- Regarding oxidised nitrogen, the analysis shows that possible acute effects are very unlikely (margin of safety estimated slightly below 1 log with 95% certainty), while chronic effects are basically non-existent.

- An attention point that needs further attention by the competent authorities is the 5-10% reduction in the dietary intake of Magnesium. This concern might however be considered more related to healthy diets rather than water safety. No studies on the exposed population have been conducted so far.

In accordance with the HACCP procedures the CCPs and POAs were identified. An overview of the CCPs and related monitoring strategies, preventive and corrective actions, is summarised in Table 5.

Table 3.8   *Identified CCPs, and related key monitoring strategies and corrective actions*

CCP	Hazard	Monitoring	Preventive action	Corrective Action
WWTP Intake	(Illegal) toxic discharge	Oxygen consumption	(Possibility to install a respirometer)	-Direct feedback -Nitrification test -discharge in canal
RO	-membrane rupture - (Involuntary) overfeeding of chemicals	Turbidity, Flow, conductivity, pH, temp., pressures, energy consumption	- Stable pretreatment (UF) operation - Cleaning procedures - Online monitoring of the chemical dosage	-Stop module - Repair O-rings - Replace broken membranes
UV	- Absorbance by shielding - Power failure	Turbidity, Flow, UV transmittance, lamp current & age	-Filter head-loss monitoring & startup procedures when they return to service -Cleaning procedures	- Check sand filter - Replace lamp
Distrib.	Recontamination	Pressure and flow changes	- maintenance of disinfectant residual -Adequate flow pressures, avoidance of sudden changes in flow rates/stagnation zones	- Isolate part of the system - Purge -Boiling alert

The scheme and procedures at Torreele/St André aquifer recharge site were already reasonably well established prior to the development of the Water Safety Plan. While there was no need to introduce new infrastructure, the process of re-evaluating the initial HACCP plan and further operational improvements brought by IWVA, identified a number of procedural adjustments, including:

- Development and testing of improved operational procedures
- Adjustment of scheme's surveillance strategy, including alarm settings
- Extension of the documentation of corrective action plans

The validation of the initial HACCP has encouraged further preventive management action, and enhanced communication channels between the wastewater (Wulpen) and drinking water (Torreele) utility. The plan helped validate security response to various scenarios which will assist in the refinement of intervention strategies, risk management success metrics and performance indicators. The WSP will be used to demonstrate due diligence for both routine and incident events.

CONCLUSIONS

As a conclusion for Atlantis, Torreele/Wulpen and Sabadell sites, it can be said from the existing data that SAT (Soil Aquifer Treatment) is a good additional barrier for treated wastewater reclamation using aquifer recharge. The aquifer acts both as passive filter, and actively through a biofilm. In this way, water is treated and some chemicals are degraded and no longer detected. This biodegradation step is especially important as compounds typically difficult to degrade may disappear (e.g. organic micro-contaminants: pharmaceuticals, estrogens, etc.). Direct injection into the aquifer (Salisbury) is also a good additional barrier, which is currently being validated on site. In the aquifer, not only dilution but a decay rate occurs for pathogens and chemical compounds.

Thus, the aquifer barrier it is an important treatment step in all the case studies, no matter the technique used to introduce the treated wastewater or stormwater into the aquifer, and the risk posed by the different hazards (microbiological, chemical and physical) is reduced. The role of the aquifer treatment is important from the health, environmental and technological points of view as well as the socio-economic one. From a marketing or acceptability perspective it can be said that water disappears from the surface which implies that treated wastewater is no longer used directly. From a legal perspective, this is of major importance since the regulations that apply to the reuse of surface and subsurface of reclaimed or reclaimed water differ considerably.

WP7: Integrative monitoring, modelling and assessment tools for subsurface treatment and storage

The four case studies investigated in WP7 cover various MAR systems in different hydrogeological and hydroclimatic contexts. The problems to be addressed through modelling for each of the investigated ReclaimWater sites are as diverse as the settings (synthesis in Table 3.9). The range of applied tools varies accordingly, from analytical solutions over 1D to 3D numerical models, taking into account conservative transport of dissolved species (salinity in particular) and pathogens as well as pathogen and contaminant attenuation, which requires combined/integrated models. Organic contaminant attenuation was addressed through the modelling of laboratory column studies of emerging pollutants (Wulpen case study), deriving biodegradation and sorption parameters, and then upscaled to field scale by 2D profile and radial numerical flow and transport models for the Wulpen and Shafdan sites. Inorganic contaminant release from the aquifer matrix was investigated for the Shafdan site through a reactive transport model taking into account the flow in the unsaturated and saturated zone (1D vertical and 1D horizontal flow), the kinetics of organic matter breakdown induced by bacterial activity in the aquifer, the resulting redox zoning in the aquifer and the specific problem of manganese release. The injection of reclaimed freshwater into a multilayer saline aquifer system was modelled with a 3D conservative transport model for the Adelaide case study, aiming to configure and optimise the ASTR system and the operating conditions so to meet the constraints on recovery salinities and residence time. Pathogen transport and attenuation in a fractured/karstified carbonate aquifer in Nardo (Italy) was simulated for a 2D set of parallel fractures allowing for pathogen deposition on fracture surfaces using a combination of an analytical approach with numerical flow modelling and particle tracking.

The **Adelaide** site (Australia) shows how modelling activities accompany all planning phases of an ASTR system in a brackish groundwater body after the initial choice and evaluation of the target aquifer. Semi-analytical and numerical (FEFLOW) finite element 2D models were used to investigate the impact of well configuration (2-6 well patterns were tested) and of well distance and injection rate on the two key regulatory constraints, maximum acceptable salinity (mixing fraction) and residence time. This allowed the design and construction of the well field that was subsequently completed by observation wells and tested through the monitoring of flushing conducted over 2 years. The acquired data served for calibrating a 3D multilayer model, a 6-layer model was found to represent best the observations. Scenarios of the evolution of mixing fractions were then calculated till 2012. Refinement of model after new data collected over the complete conditioning phase is underway and it is projected to apply ASRRI (analytical) and/or PhT3D (numerical) models to assess the fate of pathogens, trace organics and biogeochemical processes when sufficient data becomes available.
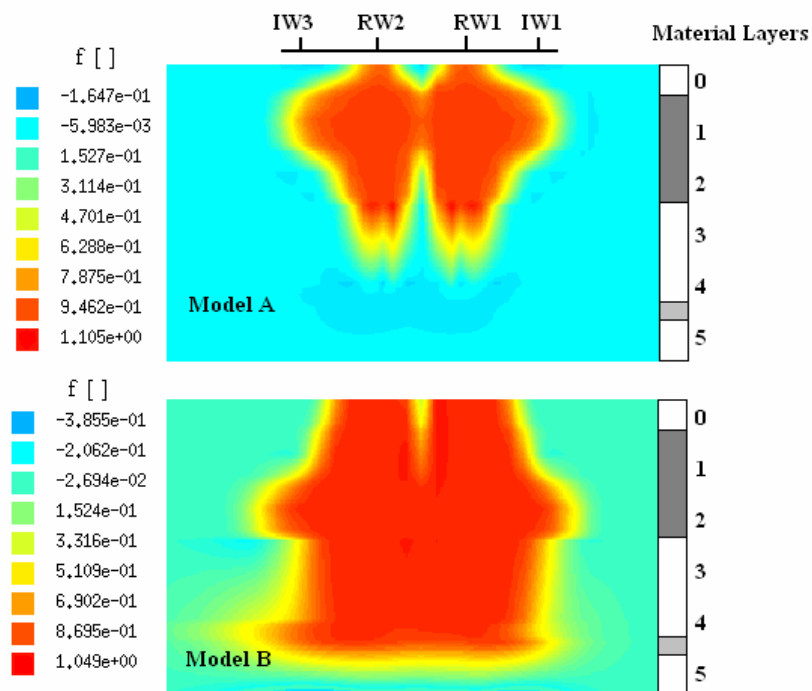
Figure 3.13 *Vertical distribution of the mixing fraction for model A and model B showing the extent of the fresh plume within the aquifer after injection through RW wells (from Kremer et al. 2008). Horizontal scale is about 250 m while vertical scale is 50 m. A and Model B and differ in terms of the degree of vertical leakage of recharge water into the underlying low permeability layers, which is significantly higher in model B, and in the porosity values with 0.7 for model A while 0.25 (more realistic than A) for model B. Both models A and B were found to fit the field data equally well but result in very different solute distribution profiles.*

At **Nardo** (Italy) the main concern is faecal contamination of a fractured/karstified carbonate aquifer. Main goal of the conducted modelling and experimental study is to estimate natural inactivation within the aquifer in function of travel time, and to evaluate the need of additional disinfection treatment of the recovered water. An analytical solution for pathogen transport in 2D parallel horizontal fractions was developed and combined with numerical particle tracking. The natural inactivation at Nardo can contribute to a single log reduction for the 5.5 d travel time between sinkhole and a 500 m distant well so that additional treatment is needed to achieve 4-logs virus reduction. Also, the minimum distance of recovery from the injection point to achieve full 4-log reduction could be estimated. Further laboratory investigations were conducted on the impact of pH, salinity, and organic compounds on virus/bacteria adsorption and disaggregation.

In **Wulpen** (Belgium), reclaimed water is treated to drinking water quality through advanced tertiary treatment before injection through infiltration ponds into a coastal dune sand aquifer and recovery by two well galleries. An important point to address with respect to regulatory constraints is the confinement of the SAT system with respect to the host aquifer. Secondly, the attenuation of trace organic compounds investigated through WP3 of ReclaimWater, was to be simulated for this system to evaluate effects of dilution-sorption-biodegradation for accidental failure scenarios.

The conclusions from a regional 3D model (using MOCDENS3D, Vandenbohede *et al.*, 2008) on the confinement of the reclaimed water within the injection-recovery zone were confirmed through a geochemical study using a novel combination of environmental isotope tracers (B, Li, O, H stable isotopes, Kloppmann *et al.*, 2008). No significant isotope fractionation was observed for the treatment process, which includes low pH RO desalination. The waste water, after infiltration trough ponds and before recovery through pumping wells is characterized by low molar Cl/B ratios (3.3 to 5.2), compared to 130 to 1020 in the wider study area, $\delta^{11}B$ values close to 0 ‰, rather homogeneous $\delta^{7}Li$ (10.3 ± 1.7 ‰) and a ^{18}O and ^{2}H enrichment with respect to ambient groundwater due to evaporation in the infiltration ponds. This confers to the AR component a unique isotopic and geochemical fingerprint. Immediately downstream of the pumping wells, and in the deeper part of the aquifer no evidence of AR wastewater could be found, indicating a high recovery efficiency. In the wider area and in the deeper part of the aquifer, isotopes evidence mixing of coastal rain and a fresh paleo-groundwater component with residual seawater as well as interaction with the aquifer material. Combining several isotope tracers provides independent constraints on groundwater flow and mixing proportions as complement to hydrodynamic modeling and geochemical studies.

The behaviour of emerging pollutants was investigated on soil columns from Wulpen (WP5) and transport/degradation parameters modelled with an 1D numerical model. In a second step, a small scale 2D model was set up to investigate tracer breakthrough on a profile from the infiltration ponds to the recovery wells and attenuation evaluated for NDMA and SMX compounds. It can be concluded that, even for the maximum observed concentrations for both compounds, concentrations at the recovery wells are not of concern.

The Shafdan case study (Israel) offers the opportunity to investigate the long-term behaviour of a large scale Soil Aquifer Treatment system over a timescale of several decades. Specific problems occurred only after 20 years of successful operation, like the reduction of infiltration capacity, partly due to physical and chemical clogging, and the sudden appearance of significant concentrations of dissolved manganese in several wells. A new injection system through dug wells was developed and tested during the project, destined to complete the existing infiltration basins, and the design and testing of the dug-well prototype was accompanied by different modelling activities. A preliminary 3D flow and conservative transport model was set up a the onset of the project to plan and design the dug well prototype and the monitoring wells. The radial pattern of solute transport obtained by the 3D model showed that a 2D radial model could satisfyingly describe the system so that a 2D flow and transport model was set up to model tracer tests and transfer scenarios of accidental peaks of organic contaminants. This model was used to simulate NDMA and SMX transfer from the injection well to the observation wells.

Boron and Li isotopes have been tested as environmental tracers of treated sewage injected into the sandy aquifer during a 38 days injection test in the newly dug injection well, a conservative artificial tracer (Br⁻) was monitored together with $\delta^{11}B$ and $\delta^{7}Li$ in the injectate, in the unsaturated soil zone (porous cup) and an observation well in the aquifer. In spite of B and Li concentrations in the injectate close to background values, significant shifts of the isotope signatures could be observed over the duration of the injection test. Boron isotope ratios show a breakthrough curve delayed with respect to Br⁻ breakthrough due to some reversible sorption on the aquifer material. No isotope fractionation was observed in the unsaturated or the saturated zone so that B-isotopes can be considered as conservative in the investigated part of the aquifer system. Lithium isotopes are strongly fractionated, probably due to sorption processes. Lithium concentrations point to a Li sink in the system, $\delta^{7}Li$ values vary strongly with a tendency of ^{7}Li depletion in the liquid phase over the duration of the

experiment. This is opposite to the expected preferential sorption of ^6Li onto clay minerals. Boron isotopes reveals a valuable tracer of artificial recharge of freshwaters derived from treated sewage, both for short term tracer tests and for long term monitoring of artificial recharge, even if in aquifers with higher clay contents, sorption-linked isotope fractionation cannot be excluded. More data are needed on Li isotope fractionation in natural groundwater systems to assess the potential of this tracer as monitoring tool.

Redox conditions were addressed through a 1D vertical unsaturated and 1D horizontal saturated reactive transport model. This model combines kinetics of microbiological degradation of organic matter (Monod kinetics), kinetic control of electron acceptors, with water-mineral interactions (dissolution-precipitation reactions). It was possible to reproduce the observed redox sequence in the Shafdan system even if this example demonstrates the limits of current bio-geochemical concepts, in particular for an element like Mn for which no stable oxidised (Mn^{IV}) aqueous species exists. Clearly, the consideration of bacterially catalysed, non-equilibrium reactions, and, simultaneously, of water-mineral equilibria through coherent bio-geochemical models is currently one of the major challenges in modelling such complex systems as are MAR systems using reclaimed water. Such models are a prerequisite for risk assessment in the field of MAR, if we want to go beyond the use of bulk parameters like sorption isotherms and biodegradation half lifes for when looking on the behaviour of organic contaminants. A thorough understanding of biogeochemical reactions occurring both in the unsaturated and the saturated zone is needed to develop predictive tools that are able to address and foresee potential drawbacks of MAR systems like pollutant breakthrough, release of trace contaminants from the aquifer material and mineral precipitations leading to a degradation of the hydraulic properties of the receiving reservoir.

Table 3.9 Overview of the ReclaimWater sites investigated through hydrodynamic (flow, transport, reactive transport) modelling

Case study	Type of reservoir	MAR system	Problems questions addressed	Modelling approach
Adelaide	Mainly porous, confined multilayer brackish aquifer	ASTR field with 2 injection and 4 recovery wells	Optimising well field design and operating conditions considering salinity and residence times.	Semianalytical and 2-3D numerical multilayer model with conservative transport.
Nardo	Fractured, karstified carbonate aquifer	ASTR with injection in a karst sinkhole and well recovery	Pathogen transport and natural attenuation in fracture system with limited filtration capacity	Analytical solution for 2D flow and transport in a series of parallel fractures combined with numerical particle tracking and flow model.
Wulpen	Coastal uncon-fined dune sand aquifer under-lain by saline groundwater			1D modelling of organic pollutant column experiments, 2D transport modelling on a profile from injection ponds to recovery wells considering sorption and biodegradation
Shafdan	Sandy unconfined aquifer with important unsaturated zone	SAT with basin infiltration, and well gallery for recovery. Pilot of dug well injection.	Design and performance evaluation of dug well injection system. Problem of redox zoning and associated Mn-release	Simple 3D model for initial screening and pilot design. 2D radial transport modelling, including unsaturated flow, sorption and biodegradation. 1Dv and 1Dh unsaturated-saturated reactive transport model including kinetic laws of bacterial growth and organic matter degradation.

4 Dissemination and use

Publishable results

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