CRANFIELD UNIVERSITY

MARIE RAFFIN

OPTIMISATION OF MEMBRANE TECHNOLOGY FOR WATER REUSE

SCHOOL OF APPLIED SCIENCES
ENVIRONMENTAL SCIENCE AND TECHNOLOGY DEPARTMENT
CRANFIELD WATER SCIENCE INSTITUTE

PhD THESIS
Academic Year: 2008 - 2011

Supervisor: Prof. Simon Judd
November 2011

CRANFIELD UNIVERSITY

SCHOOL OF APPLIED SCIENCES
ENVIRONMENTAL SCIENCE AND TECHNOLOGY DEPARTMENT
CRANFIELD WATER SCIENCE INSTITUTE

PhD THESIS

Academic Year 2008 - 2011

MARIE RAFFIN

OPTIMISATION OF MEMBRANE TECHNOLOGY FOR WATER REUSE

Supervisor: Prof. Simon Judd

November 2011

This thesis is submitted in partial fulfilment of the requirements for the degree of Doctor of Philosophy

© Cranfield University 2011. All rights reserved. No part of this publication may be reproduced without the written permission of the copyright owner.

ABSTRACT

Increasing freshwater scarcity is making reclamation of wastewater effluent more economically attractive as a means of preserving freshwater resources. The use of an integrated membrane system (IMS), the combination of micro/ultra-filtration (MF/UF) followed by reverse osmosis (RO) membranes, represents a key process for municipal wastewater reuse.

A major drawback of such systems is the fouling of both the MF/UF and RO membranes. The water to be treated by the IMS system varies from one wastewater treatment plant (WWTP) to another, and its fouling propensity changes correspondingly. It is thus preferable to conduct pilot trials before implementing a full-scale plant. This thesis aims to look at the sustainability of IMS technology dedicated to indirect potable reuse (IPR) in terms of fouling minimisation and cost via a 600 $m^3.d^{-1}$ pilot plant.

Wastewater reuse plants, using IMS, as well as statistical methods for membrane optimisation were reviewed. Box-Behnken design was used to define optimum operating envelopes of the pilot plant for both the microfiltration and the reverse osmosis in terms of fouling minimisation. Same statistical method was used to enhance the efficiency of the MF cleaning-in place through bench-scale test.

Data from the pilot plant MF process allow to determine relationship between reversible and irreversible fouling, and operating parameters and feed water quality.

Life cycle cost analysis (LCCA) of the both trains (MF/RO/AOP and MF/AOP) of the pilot plant was performed and compared with the LCCA of two full-scale plant.

Keywords: Integrated membrane system; wastewater reuse; optimisation; Box-Behnken design; operating parameters; fouling; Life cycle cost.

ACKNOWLEDGEMENTS

Firstly, I wish to thank Thames Water who fully funded this research and gave me the opportunity to work on a really interesting project.

Sincere thanks to Simon Judd, without whom this thesis would not be what it is. Many thanks for his support and his advice during these three years and especially his patience while trying to understand my French-English.

I would also like to thank Eve Germain, my industrial supervisor, who was always here to advise me and answer to my questions. Thanks to everyone at Thames Water Innovation, particularly Martyn, Chris, Juliette, Pete and Gemma from the IPR pilot plant, Andrew, Sonia and Bart. My three years at the pilot plant would not have been the same without you. I would also like to thank Sian Hills and Angela Barugh, it was a pleasure to work with you.

I would like to thank Rosa Daviu-Castello and Aqil Shiffar for their help. You saved me a lot of time and free me from unnecessary stress.

On a technical side, I would like to thanks Alastair Halliday, from Thames Water, who helped me on the comprehension of statistical methods and all the people from Thames Water laboratory for all the analyses they have done for me. Thanks to Thomas Koane-Yane who provided me with a macro, transforming 6 months of manual data analysis to 1 month of automatic data analysis.

Last but not least, I would like to thanks my family, especially my parents and my brothers, who supported me all over these years, and my friends either in England or in France.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

LIST OF ABBREVIATIONS

AAN	Artificial neural network
ANOVA	Analysis of variance
AOP	Advanced oxidation process
BBD	Box-Behnken design
BSA	Bovine serum albumin
BW	Backwash
C	Chemical concentration
CAPEX	Capital Expenditure
CCD	Central composite design
CEB	Chemically-enhanced backwash
CIP	Cleaning in place
COD	Chemical oxygen demand
DOC	Dissolved organic carbon
EPS	Extracellular polymeric substances
ICP-OES	Inductively-coupled plasma-optical emission spectrometer
IMS	Integrated membrane system
IPR	Indirect potable reuse
J	Permeate flux
K	Membrane permeability
K_f	Permeability after CIP
K_i	Permeability before CIP
$K_{irreversible}$	Irreversible fouling rate
$K_{reversible}$	Reversible fouling rate
LCC	Life cycle cost
LCCA	Life cycle analysis
LMH	Litre per square meter per hour
LSI	Langelier saturation index
MBR	Membrane bioreactor
MF	Microfiltration
MLD	Mega litre per day
NOM	Natural organic matter
OPEX	Operational expenditure
O&M	Operation and maintenance

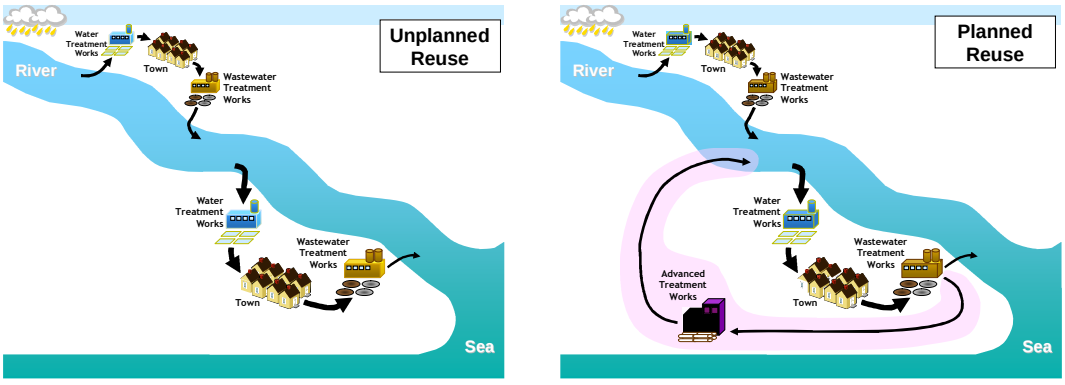
PA	Polyamide
PES	Polyethersulfone
pH_S	Saturation pH
PVDF	Polyvinylidene Difluoride
R^2	Coefficient of determination
RO	Reverse osmosis
RSM	Response surface methodology
S	Soaking time
SCADA	Supervisory control and data acquisition
SDI	Silt density index
SED	Specific energy demand
SEM-EXD	Scan electron microscopy – Energy dispersive X-ray
SUVA	Specific UV absorbance
T	Temperature
TDS	Total dissolved solids
TMP	Transmembrane pressure
TMP_0	Initial TMP
TMP_{max}	Maximum transmembrane pressure of operation
TOC	Total organic carbon
TSS	Total suspended solids
UF	Ultrafiltration
UV	Ultraviolet
WWTP	Wastewater treatment plant
Y_{exp}	Experimental volumetric ratio
Y_{model}	Volumetric ratio defined by the model equation
$\%R_{exp}$	Experimental permeability recovery
$\%R_{predicted}$	Permeability recovery predicted by the model

CHAPTER 1: INTRODUCTION

1.1 Background

Increasing freshwater scarcity is making reclamation of wastewater effluent more economically attractive as a means of preserving freshwater resources. Whilst the conventional solution to freshwater resourcing in arid regions has been to desalinate seawater, it is widely recognised that reuse is more energetically efficient even when employing membrane technology to provide the same high-quality permeate product (Markus and Deshmukh, 2010; Rodriguez *et al.*, 2009).

Currently, and commonly in most urban areas, unplanned wastewater reuse already takes place: water may be extracted from the water body (such as a river) downstream of the discharge point of a wastewater treatment plant (WWTP) in an unregulated manner (Figure 1-1 (a)). The idea of planned wastewater reuse is to retain part of the WWTP effluent and treat it further via an advanced treatment plant, in which the goal is to achieve a water quality target to allow it to be safely reused. Reclaimed water from the advanced treatment plant has a number of applications, including industrial process water (Macbeth *et al.*, 2004), indirect potable reuse (Van Houtte and Verbauwhede, 2008), direct potable reuse (Du Pisani, 2006), conservation and increase of environmental flow (Esteban and Ortega de Miguel, 2008), barrier against seawater intrusion (Cazurra, 2008), non-potable municipal reuse (Lazarova *et al.*, 2003) and irrigation (Gomez Gotor *et al.*, 2001).



(a) (b)

Figure 1-1 Unplanned reuse (a) and planned reuse of wastewater (IPR) (b)

In the case of indirect potable reuse (IPR) (Figure 1-1(b)), reclaimed water is injected in the catchment of a drinking water plant, which can be a reservoir (Freeman *et al.*, 2008), an aquifer (Markus and Deshmukh, 2010) or river (e.g. Essex and Suffolk advanced treatment plant). Planned IPR already exists and is mainly located in USA, Singapore and Australia (Figure 1-2). Most of these plants are membrane-based and are either "polishing" systems, whereby wastewater already treated by a conventional wastewater treatment plant (WWTP) is further treated to potable water quality or better, or "total" systems where the raw sewage is treated directly. While membrane bioreactor (MBR) technology is increasingly used for the latter, there remains a significant number of polishing plants based on a combination of either microfiltration (MF) or ultrafiltration (UF), with or without downstream reverse osmosis (RO), with other upstream and/or downstream processes also demanded depending on circumstances.

A major drawback of such systems is the fouling of both the MF/UF and RO membranes, also known as integrated membrane system (IMS). Membrane fouling reduces the throughput of the process as well as increasing the cost. The water to be treated by the IMS system varies from one WWTP to another, and its fouling propensity changes correspondingly. It is thus preferable to conduct pilot trials before implementing a full-scale plant.

With 613 mm/year of average rainfall, London can be seen as a "cloudy" desert whilst its water demand is inexorably increasing with increasing population and economic growth. To increase its drinking water supply, the regional water utility, Thames Water, is investigating the feasibility of the use of "planned" indirect potable reuse (IPR) by implementing a 600 m^3/d pilot plant in North London. The pilot plant was designed based on existing worldwide schemes using state-of-the art technology at the time, with a "multi-barrier" approach using of MF, RO and an advanced oxidation process (AOP).

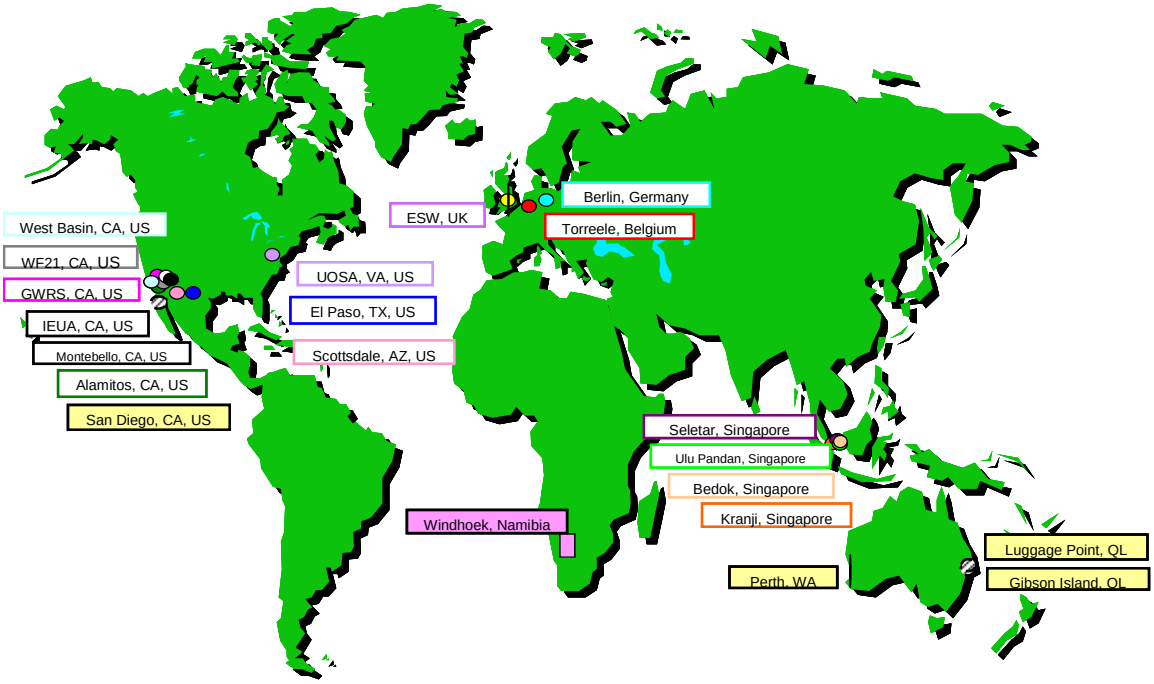


Figure 1-2 IPR plants in the world

1.2 Aims and objectives

The present thesis reports the results of a three-year research study which was fully-funded by Thames Water. This thesis aims analyse the sustainability of IMS technology dedicated to IPR in terms of fouling minimisation and cost. The objectives principally comprised:

1. appraisal of current practice for wastewater reclamation using MF/UF-RO treatment process,
2. assessment of statistical experimental programming for membrane optimisation, in particular Box-Behnken design,
3. optimisation of both processes in terms of operating parameters for minimising fouling and scaling of the membrane, and specifically
4. optimisation of the MF process in terms of flux, backwash frequency, chloramine dose, chemical cleaning protocol and feed water quality,
5. optimisation of the RO process in terms of flux, recovery, antiscalant dose and type and pH, and

6. assessment of life cycle cost (LCC) of such a plant, including the influence of operating parameters on the LCC.

Although this work is based on membrane fouling, the topics of membrane fouling characterisation and mechanisms are not reviewed since these are extremely well explored in a number of review articles and reference books. Table 1-1 privdes an overview of recent membrane fouling review publications in learned journals and books for both MF/UF and RO membranes.

Table 1-1 Recent publications on membrane fouling

	Process	*References*
Books		Nath, K. (2008), *Membrane Separation Processes*, Prentice-Hall of India, New Dehli Wilf, M. (2010), *Membrane Technology for wastewater reclamation*, Balaban Desalination Publications, Hopkinton, USA Judd, S., Jefferson, B. (2003), *Membranes for Industrial wastewater recovery and re-use*, Elsevier LTD, Oxford, UK
Review	UF	Gao, W., Liang, H., Ma, J., Han, M., Chen, Z.-L., Han, Z.-S., Li, G.-B. (2011), Membrane fouling control in ultrafiltration technology for drinking water production: A review, *Desalination*, 272 (1-3), p. 1-8
	RO/UF	Goosen, M.F.A., Sablani, S.S., Ai-Hinai, H., Ai-Obeidani, S., Al-Belushi, R., Jackson, D. (2004), Fouling of reverse osmosis and ultrafiltration membranes: a critical review, Separation and Science Technology, 39, p.2261–2297
	RO	Tang, C.Y., Chong, T.H., Fane, A.G. (2011), Colloidal interactions and fouling of NF and RO membranes: A review, *Advances in Colloid and Interface Science,* 164 (1-2), p. 126-143

1.3 Thesis structure

This thesis is presented in paper format. Apart from Chapter 3, which was written by Rosa Daviu Castello (MSc student from Cranfield University) as part of her MSc thesis and who supervised by the author, Marie Raffin, all papers were written by the author, with Professor Simon Judd acting as corresponding

author on submitted journal papers. All the experimental work was undertaken by the author.

A review of existing wastewater reuse plants using integrated membrane (MF/UF-RO) system, based on published literature and a bespoke survey of IMS installations, is provided in Chapter 2. This review looked at the pre-treatment of the IMS, the operating parameters applied and the costs involved. (Submitted for Publication to *Environmental Technology*: Wastewater reuse using MF/UF - RO processes: a review of existing installations, Raffin, M., Shiffar, A., Germain, E., Judd, S.).

Chapter 3 provides an outline review of existing statistical experimental programming as applied generally and to membrane process optimisation specifically. Comparison with methods identified with Box-Behnken design (BBD) is made with specific reference to the number of experiments, the simplicity of the calculation, the order of the response, the estimation of error and the distribution of information throughout the region of interest. It was concluded that BBD appears to offer an appropriate and efficient method for experimental design for optimising membrane processes. (Submitted for publication to *Membranes*: Daviu, R., Raffin, M., Germain, E. and Judd., S.,Statistical experimental programming for membrane process optimisation).

Chapter 4 presents the results of the optimisation of both the MF and the RO process in terms of fouling/scaling minimisation. An envelope of optimum operating parameters has been defined for both processes, as a function of flux and backwash frequency for the MF and recovery and pH for the RO. (Published in *Desalination* 273 (2011), p. 136–141: Raffin, M., Germain, E. and Judd, S., Optimising operation of an integrated membrane system (IMS) – A Box-Behnken approach).

Chapter 5 presents the results of an extensive study of the influence of operating parameters (such as flux and backwash interval) and feed water quality parameters (turbidity and temperature) on reversible and irreversible fouling rate, from which the cleaning-in-place (CIP) interval is defined.

(Submitted to *Water Research*: Raffin, M., Germain, E. and Judd, S., Backwash and flux optimisation for indirect potable reuse using microfiltration)

Chapter 6 provides a study of the optimisation of the microfiltration process with specific reference to the CIP. BBD was used to optimise the chemical clean as a function of the chemical cleaning reagent, its concentration, and the soak time and temperature. (Published in *Separation and Purification Technology* 80 (2011), p. 452–458: Raffin, M., Germain, E. and Judd, S., Optimisation of MF membrane cleaning protocol in an Indirect Potable Reuse (IPR) scheme)

Assessment of fouling of the RO process is reported in Chapter 7 and includes the membrane autopsy of three RO modules, along with a cursory study of the efficiency of different antiscalants. (Submitted to *Desalination and Water Treatment*: Raffin, M., Germain, E. and Judd, S., Assessment of fouling of an RO process dedicated to indirect potable reuse)

Chapter 8 presents the results of the life cycle cost analysis of the pilot plant along with those projected for two full-scale plants. Assumed operating conditions for the full-scale plants were the same as those identified for the pilot plant. (To be submitted to *Desalination*: Raffin, M., Germain, E. and Judd, S., Life Cycle Cost Analysis (LCCA) for an Indirect Potable Reuse Scheme – From pilot plant to potential full-scale plant)

The conclusions are summarised in Chapter 9, where suggestions for future work are also provided. This chapter crystallises the key outcomes of the work, which can be depicted in the form of a road map (Figure 1-3) which illustrates how the various topics correlate.

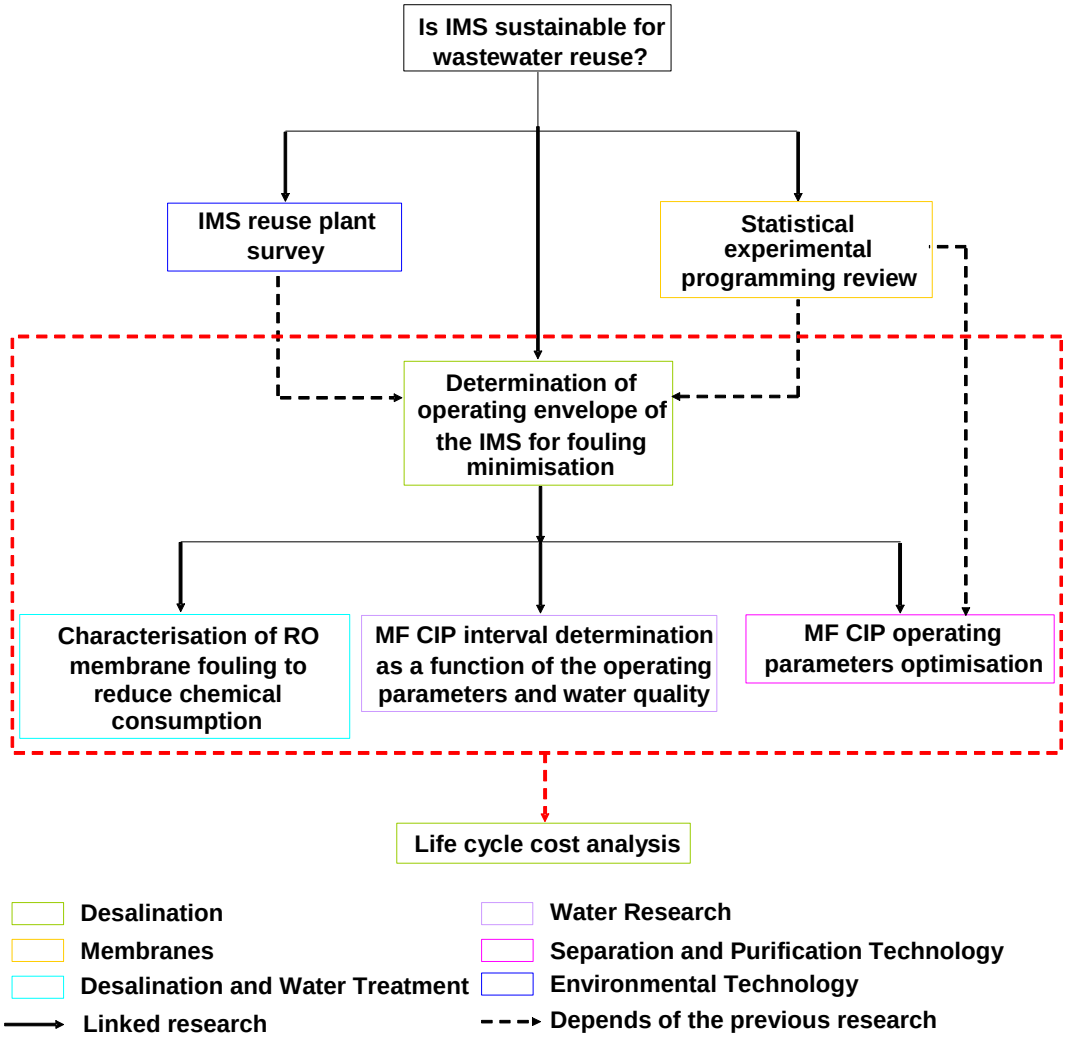


Figure 1-3 Thesis roadmap

1.4 References

Cazurra, T. (2008), Water reuse of south Barcelona's wastewater reclamation plant, *Desalination*, 218, p. 43-51.

Du Pisani, P. L. (2006), Direct reclamation of potable water at Windhoek's Goreangab reclamation plant, *Desalination,* 188 (1-3), p. 79-88.

Esteban R. I., Ortega de Miguel, E. (2008), Present and future of wastewater reuse in Spain, *Desalination*, 218, p. 105-119.

Freeman, S., Bates, J., Wallis-Lage, C., McEvoy, J. (2008), Drought relief in South East, Queensland, Australia, provided by membrane-reclaimed water, *AWWA Journal*, 100 (2), p. 40-52.

Gómez Gotor, A., Pérez Baez, S.O., Espinoza, C.A., Bachir, S.I. (2001), Membrane processes for the recovery and reuse of wastewater in agriculture, *Desalination*, 137, p. 187-192.

Lazarova, V., Sheilds, P., Levine, B., Savoye, P., Hranisavljevic, D., Renaud, P. (2003), Production of high quality for reuse purposes: the West Basin experience, *Water Science and Technology: Water Supply*, 3 (3), p. 167-175.

MacBeth, I., Murrer, J., Latter, S. (2004), Reducing the costs for ultra-pure water production for use as de-min plant feed water for Peterborough power station, UK, Chemie im Kraftwerk 2004.

Markus, M.R., Deshmukh, S.S. (2010), An innovative approach to water supply - The Groundwater Replenishment System, In: *World Environmental and Water Ressources Congress 2010: Challenges of Change*, Providence, Rhode Island 16-20 May 2010, p. 3624-3639.

Rodriguez, D., Van Buynder, P., Lugg, R., Blair, P., Devine, B., Cook, A. and Weinstein, P. (2009), Indirect potable reuse: A sustainable water supply alternative, *International Journal of Environmental Research and Public Health,* 6 (3), p. 1174-1209.

Van Houtte, E., Verbauwhede, J. (2008) Operational experience with indirect potable reuse at the Flemish Coast, *Desalination*, 218, p. 198–207.

CHAPTER 2: WASTEWATER REUSE USING MF/UF-RO PROCESSES: A REVIEW OF EXISTING INSTALLATIONS

Raffin, M., Shiffar, A., Germain, E., Judd, S., Wastewater reuse using MF/UF-RO processes: a review of existing installations, submitted to *Environmental Technology*

2.1 Introduction

The increased global implementation of planned wastewater reuse, in which the goal is to treat the water to a level where it may be safely reused, has arisen from commensurately increased stresses on freshwater supply. This is to be differentiated from unplanned wastewater reuse, where water may be extracted from the water body (such as a river) downstream of the discharge point in an unregulated manner. Whilst the conventional solution to freshwater resourcing in arid regions has been to desalinate seawater, it is widely recognised that reuse is more energetically efficient even when employing membrane technology to provide the same high-quality permeate product (Markus and Deshmukh, 2010; Rodriguez *et al.*, 2009).

There are an increasing number of wastewater reuse installations worldwide based on membrane technology, providing an absolute barrier to potentially harmful pathogenic microorganisms. These installations may be either be “polishing” systems, whereby wastewater already treated by a conventional wastewater treatment plant (WWTP) is further treated to potable water quality or better, or “total” systems where the raw sewage is treated directly. The latter is appropriate for a green-field site, and invariably employ membrane bioreactor (MBR) technology. Polishing plants, on the other hand, are well established and are appropriate when an existing conventional WWTP requires upgrading to provide reusable water. While MBR technology is increasingly used, with the largest MBR plant of 495 MLD planned for Brightwater, WA, to be commissioned in early 2012, there remains a significant number of polishing plants, all of these based on a combination of either microfiltration (MF) or ultrafiltration (UF) with or without downstream reverse osmosis (RO), with other upstream and/or downstream processes also demanded depending on circumstances. These installations are amongst the largest membrane plants in the world (Table 2-1). Among the ten largest membrane based wastewater reuse plant, six are based on MF/UF alone with no downstream RO. These plants provide water for irrigation, requiring a lower effluent water quality. RO is used when a high product water quality is needed, such as for reuse as boiler feedwater or planned indirect potable reuse.

Table 2-1 Ten largest membrane-based wastewater reuse plants worldwide, April 2012

Site	*Membrane*		*Applications*	*Commissioned*	*Capacity (MLD)*
	MF/UF	*RO*			
Doha North, Qatar	Norit	-	Irrigation	2011	440
Sulaibiya, Kuwait	Norit	Toray	Irrigation	2004	375
Orange County,USA	Siemens/Memcor	Hydranautics	Groundwater replenishment	2008	328
Changi, Singapore	Siemens/Memcor	Toray	Industry, indirect potable reuse	2010	232
Ulu Pandan, Singapore	Pall/Asahi	Hydranautics	Industry, indirect potable reuse	2007	191
Gwinnet County,GA,USA	GE/ZENON	-	Irrigation	2005	289
Doha South, Qatar	Norit	-	Irrigation	2012	187
Qinghe Phase II China	Norit	-	Industry, Irrigation, Municipal non-potable reuse	2010	180
Agra, India	Siemens/Memcor	-	-	2010	144
Doha west, Qatar	GE/ZENON	-	Irrigation	2009	135

In this paper, nine existing membrane installations based on MF/UF-RO systems, also known as integrated membrane system (IMS), are reviewed. Their performance is appraised to ascertain any trends in operation and maintenance, hydraulic performance (i.e. membrane permeability) and water quality, cost and operability, since their economic viability is highly dependent on these facets.

2.2 Methodology

2.2.1 Literature survey

A survey was performed to obtain information on existing IMS wastewater installations worldwide. Information sources included peer-reviewed journals (identified via database searches using *Scopus*), conference proceedings, supplier websites, key reference texts (Wilf, 2010; Jimenez and Asano, 2008; Asano, 2006) and personal contacts. A number of plants were selected for further examination from those originally identified (Appendix 1) on the basis of comprehensiveness of the available information.

2.2.2 Plant survey

Operational and maintenance data were acquired from a survey completed in the period between October 2009 and April 2011. The survey aimed to acquire water quality and key technical data relating to the operation and maintenance (O&M) of the wastewater reuse installations. A template was developed (Appendix 2), adapted from that used for previous similar surveys of MBR installations (Judd and Judd, 2010). 15 plants were originally targeted, each with identified named individuals as survey recipients. Of these 15 installations, following further contact with the survey recipients for clarification, sufficiently comprehensive information was acquired from nine sites (Table 2-2).

The sites surveyed had design flows ranging from 1.6 to 375 MLD, including some of the largest reuse plants in the world as well as some smaller-scale well-established installations, from across the USA, Europe, the Middle East, South-East Asia and Australia. Of these installations, the largest was based on the Norit technology, and it is this technology which also provides four of the largest installations globally (Table 2-1). All identified plants treat secondary municipal effluent, both with and without nutrient removal.

Table 2-2 Installation surveyed

Site	*Location*	*MF/UF supplier*	*RO supplier*	*Capacity (MLD)*
Plant A	UK	Asahi Kasei/Pall	Koch	1.6
Plant B	Australia	Asahi Kasei/Pall	Toray	66
Plant C	USA	Asahi Kasei/Pall	Hydranautics	11.4
Plant D	Singapore	Asahi Kasei/Pall	Hydranautics	191
Plant E	Singapore	Siemens/Memcor	Toray	232
Plant F	Spain	GE/Zenon	DOW	15
Plant G	Kuwait	Norit	Toray	375
Plant H	Belgium	GE/Zenon	DOW	6.9
Plant I	USA	Siemens/Memcor	Hydranautics	265

2.3 Results

2.3.1 Current status

Membrane-based wastewater reuse began in 1975 with the start up of Water Factory 21, based on conventional pre-treatment followed by RO. However, it was only from the mid 1990s that the cumulative installed capacity for wastewater reuse began to increase (Figure 2-1) as membrane costs concomitantly decreased (Jud and Judd, 2010). Most of the plants are located in USA (11 plants), Australia (6 plants) and Singapore (5 plants), representing 76% of the global number of plants. The USA and Singapore each provide ~31% of the total reclaimed water produced worldwide each year. The largest membrane-based municipal wastewater reuse plant, however, is in Kuwait: the 320 MLD plant at Plant G provides ~20% of the world's IMS wastewater (Figure 2-2). The recovered water has a number of applications including industrial process water (Macbeth *et al.*, 2004), indirect potable reuse (Van Houtte and Verbauwhede, 2008) via a reservoir (Freeman *et al.*, 2008), groundwater recharge

(Markus and Deshmukh, 2010) or river (e.g. Essex and Suffolk advanced treatment plant),direct potable reuse (Du Pisani, 2006), conservation and increase of environmental flow (Esteban and Ortega de Miguel, 2008), barrier against seawater intrusion (Cazurra, 2008), non-potable municipal reuse (Lazarova *et al.*, 2003) and irrigation (Gomez Gotor *et al.*, 2001). For the latter two categories lower tech process are generally preferred on the basis of cost.

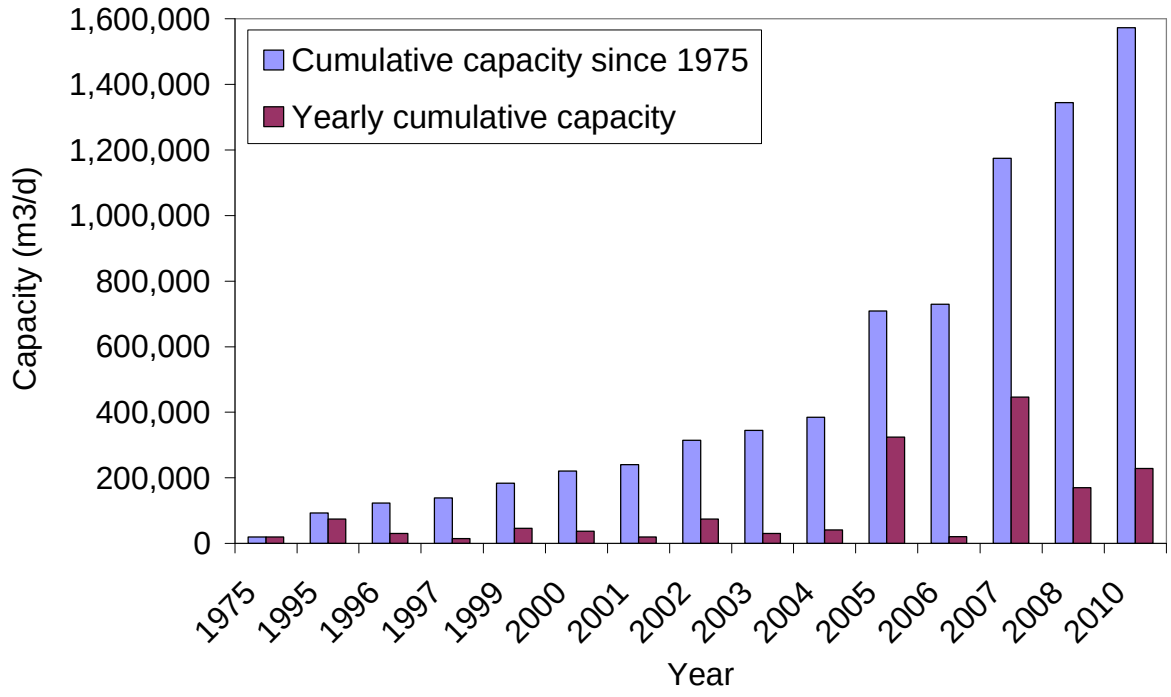


Figure 2-1 Total and yearly cumulative capacity of IMS wastewater plants (from Appendix 1)

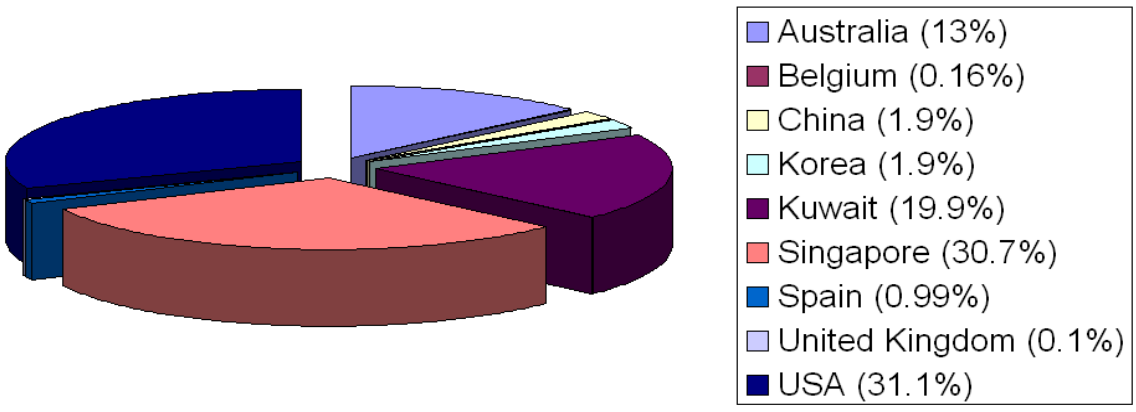


Figure 2-2 IMS Wastewater plants capacity (%) per country (from Appendix 1)

2.3.2 Technology

2.3.2.1 Pre-treatment

Pre-treatment of the secondary/tertiary effluent by chemical dosing and/or straining is normally necessary to protect the MF/UF from fouling, clogging and possible physical damage from extraneous particles. Of the nine installations surveyed, screening was used for 8 plants with mesh sizes ranging from 0.01 to 2 mm; the tendency appears to be to have a lower mesh size with a higher feed water turbidity and suspended solids concentration (Figure 2-3). However, screens have been shown to be susceptible to clogging by large solids coupled with tenacious biological growth demanding rigorous backwashing (Hatt et al, 2010).

Coagulation using either PAX or ferric-based coagulant is used at three of the plants for removing phosphate and/or decreasing total organic carbon (TOC) concentration, and thus reduce fouling on both MF and RO membranes by suppressing biofouling, organic fouling and scaling. In the surveyed plants, coagulant was dosed either before the screen, upstream of flocculation and clarification, or upstream of the MF membrane, with the latter two options being the most common. It is recognised that precoagulation of dissolved organic matter upstream of MF/UF membranes reduces fouling as well as enhancing removal of organic material that otherwise is largely unremoved by microporous membranes (Jung et. al 2006). Against this, overdosing with coagulant may exacerbate membrane fouling, especially for the RO membrane (Gabelich *et al.*, 2006; Moon *et al.*, 2009).

Chloramination/chlorination was used at 6 plants, ostensibly to suppress biofouling of the membrane by inactivating micro-organisms and oxidising organic material. Chloramination is only mildly oxidising but a reasonably effective biocide which does not cause damage to the downstream RO membrane. Chloramines maybe be either pre-formed or generated in situ from sodium hypochlorite and ammonium sulphate. The use of chlorination demands that sodium bisulphate is dosed prior the RO to quench the chlorine, since RO membranes are generally chlorine intolerant (Soice *et al.*, 2003).

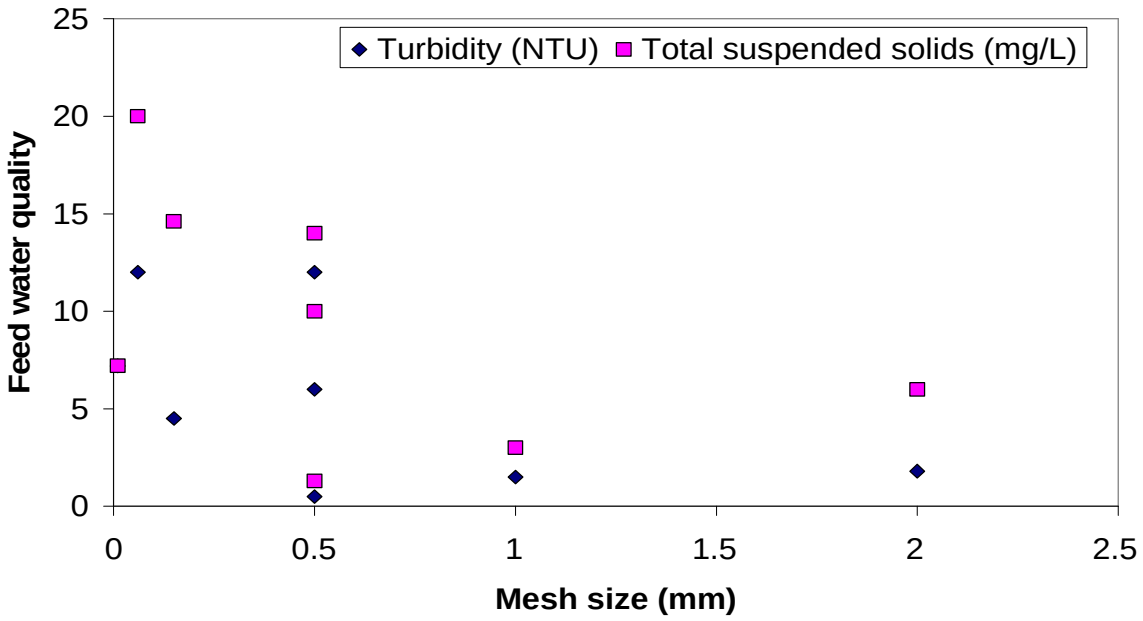


Figure 2-3 Feed water quality (turbidity and Total suspended solids) of the pre-treatment as a function of the screening mesh size (mm)

2.3.2.2 Microfiltration / Ultrafiltration

2.3.2.2.1 Membrane type (Table 2-3)

MF/UF membrane suppliers comprise Pall/Asahi Kasei (4 sites), Siemens/Memcor (2 sites), GE/Zenon (2 sites), and Norit (1 site). The principal membrane properties are given in Table 2-3. These include the membrane configuration (either hollow fibre (HF) or capillary tube (CT) depending respectively on whether flow is from outside to inside the lumen or vice versa), membrane pore size, and module configuration (either pressurised on the feed side or else submerged in a tank and the permeate withdrawn under suction). Of the nine sites reviewed, four are fitted with MF membranes (pore size above 0.08 µm) with the remainder being UF. The membrane pore size has no impact on flux, which instead depends on the membrane configuration; the pressurised CT membranes operate at a substantially higher flux than the pressured HF products. The immersed membranes operate at the lowest flux, since these rely on applying a partial vacuum on the permeate side.

2.3.2.2.2 Flux, permeability and conversion (Table 2-3)

The MF/UF design flux values across all plants range from 28 to 65 LMH, with an average of 46 LMH - or 52 LMH after temperature-correcting to 20°C (Table 2-5). Elevated fluxes would be expected to relate either to the CT membrane or from a less highly-fouling feedwater. Such operating conditions would otherwise incur more excessive fouling and, consequently, more frequent membrane cleaning. Trans-membrane pressure (TMP) ranges are reasonably consistent across all sites, averaging 0.35 bar. The only outlier is Plant E, which has a reported TMP value of 0.8 bar and a corresponding low permeability (flux/TMP ratio). Permeability values otherwise exceed 150 LMH/bar, with no recognisable trend with either membrane or module configuration. Conversions are also reasonably consistent, ranging from 88 to 95% and reflecting small losses of product water used for periodic backwashing of the membrane to recover permeability

2.3.2.2.3 Membrane cleaning

The key remaining O&M parameters are those relating to membrane cleaning. HF and CP membranes are normally cleaned by backwashing and chemically cleaning (Table 2-6).

Of the nine sites, seven employ an air and water backwash, where air is applied to the outside of the fibres to scour the feed side of the membrane and a small amount of filtrate is reversed through the membrane. The two other plants used only water, although for the Zenon process air is used intermittently to create turbulence along the membrane length (25% of the filtration time). Backwash intervals varied between 8 minutes to 38 minutes with an average of 23 min. It would be expected that the backwash interval would depend on the flux applied, membrane configuration or water quality such as turbidity and total suspended solids (TSS) since such parameters influence permeability decline rate (Liu *et al.*, 2009; Wang *et al.*, 2008; Citulski et al, 2009). However, it appears that, apart from one outlier, the stronger correlation is with water temperature (Figure 2-4), as recognised in previously reported studies (Wang, 1988).

Backwashing is known to remove primarily reversible fouling (Psoch and Schiewer, 2006), whereas supplementary periodic chemical cleaning is required to remove

irreversible fouling (Kimura *et al.*, 2006). Chemical cleaning is normally achieved without removing the membranes (hence clean in place or CIP), and cleans are often applied which combine backwashing with chemical cleaning (a chemically-enhanced backwash or CEB). The frequency of cleans and chemical dosing requirements (Table 2-6) reveal MF/UF CIP frequency to be conducted once or twice monthly in most cases, with only the CT-based plant operating with apparently just a yearly chemical clean. The high feedwater COD demands that Plant D plant requires a chemically-enhanced backwash (CEB) up to three times a day (Table 2-6). Chemical cleaning demand is increased at high fluxes, as reflected in the cleaning requirements of Plants G and B. Plant H demands a CEB every 25 backwash cycles, which could be linked with the coarse screening rather than fine screening pre-treatment. Sodium hypochlorite is widely used for chemical cleaning by both CIP and CEB and may be mixed with NaOH to enhance cleaning if the membranes are tolerant to high pH level. Chelating acid such as citric acid and oxalic acid are used to supplement the hypochlorite clean. This hypochlorite/organic acid combination is common in cleaning of membrane municipal plants.

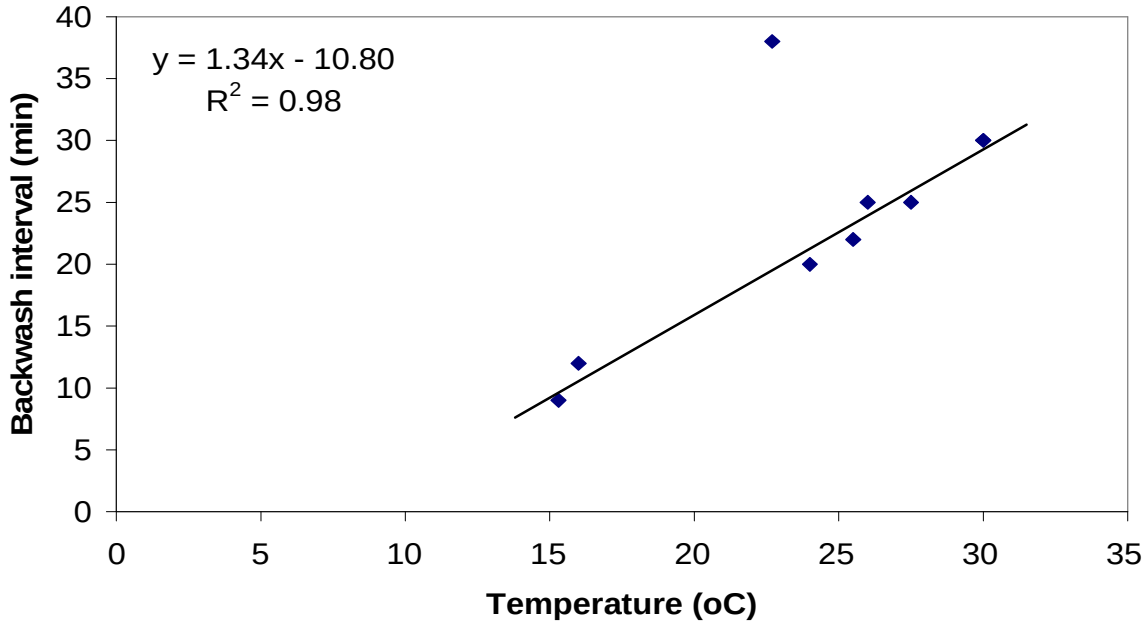


Figure 2-4 Backwash interval as a function of the water temperature

2.3.2.3 Reverse osmosis

2.3.2.3.1 Membrane type (Table 2-3)

RO membrane suppliers comprise Toray (3 sites), Dow (2 sites), Hydranautics (2 sites), Koch (1 site), and Hydranautics (1 site). Current commercialised membranes are exclusively spiral wound in configuration, and these are not backwashable. Cross-flow filtration is used to provide membrane scouring, generating a waste concentrate flow and so limiting the conversion from a single stage. More than one stage is thus normally required, forming an "array", to achieve adequate overall conversion (Judd and Jefferson, 2003).

2.3.2.3.2 Flux and conversion (Table 2-3)

Data from the RO process indicate a fairly consistent range of fluxes (Table 2-3), with a mean value of 19.3 LMH. The RO process employed across the sites is mainly as a two-stage array (7 plants) providing 75-80% conversion, as opposed to the three stage arrays (2 plants) which provide ~85% conversion.

The conversion per element θ_e is calculated from the permeate to feed flow ratio:

$$\frac{Q_p}{Q_f} = 1 - (1 - \theta_e)_n$$

where *n* the number of elements across the array. It follows that for an individual stage where each module has *m* elements, the conversion is given by:

$$\theta_{stage} = 1 - (1 - \theta_e)^m$$

Applying either of the above equations, the calculated number of elements per pressure vessel is 6 or 7 across all sites. For an assumed 50% conversion per stage, as would be normal, the mean conversion per element is 9.4-10.9%. This is within the overall range of conversion per element of 8.6 to 12.7% (Table 2-7).

Lower fluxes arise for the plants in Singapore as well as for Plant C, respectively 18 LMH for Plant D and 17 LMH for Plant E and Plant C. For the Singapore plant, it might be assumed that the flux and the recovery were based on the experience at Bedok NEWater demonstration plant, where organic fouling and calcium phosphate scaling occured on the 2nd stage at the start up of the plant (Bartels *et al.*, 2007).

Looking at water quality data for both plants, it appears likely that Plants D and E may also be susceptible to such fouling, explaining the rather conservative flux.

For Plant C, a conversion of 85% is applied, significantly increasing fouling potential. Such a high conversion is generally used when a low concentrate flow is demanded. When comparing the results from the two plants at 85% recovery, Plant I (20 LMH flux) and Plant C, it appears clearly that the difference between both plants is the water quality upstream of the RO. Although the TOC is higher at Plant I, the SDI and phosphate concentration are higher than that at Plant I leading to higher fouling potential by particle and colloidal matter as well as increase phosphate scaling potential (Xu *et al.*, 2010; Greenberg *et al.*, 2005).

The fouling nature of water is normally determined by the Silt Density Index (SDI), which is a measure of the rate at which membrane pores plug and must normally be below 3 for operation of an RO process. Since fouling increases with the SDI, higher SDI waters may be expected to correlate with either a decreased flux or an increased cleaning (i.e. CIP) frequency. Figure 2-5 indicates an approximate linear relationship between SDI and CIP frequency. The exceptional datum (SDI 4, CIP frequency 6/year) corresponds to Plant C. While SDI provides a useful guide to pore plugging propensity, it provides no information regarding precipitation of sparingly soluble salts; the anomalous datum for Plant C may arise from phosphate scaling rather than membrane pore plugging.

Scaling may be reduced by the use of chemical pre-treatment such as acid and antiscalant. Of the nine plants studied, four use acid dosing with antiscalant and three use antiscalant alone; information for the remaining plants was unavailable. pH correction leads to relatively low pH levels of 6.8 or less. Contrary to expectation, there is no acid dosing with the three-stage RO process which yields higher conversions. Plant F, with low conversion, apparently operates without acid dosing.

2.3.2.3.3 Membrane cleaning (Table 2-6)

Compared with the MF/UF the RO chemical cleaning requirements are moderate. The CIP frequency across all plants ranges from 2 to 6 times per year (Table 2-6), the cleaning demand relating to the water quality (Figure 2-5) and the operating parameters regarding scaling minimisation.

Table 2-3 Key O&M parameters

Site:	*Plant A*	*Plant B*	*Plant C*	*Plant D*	*Plant E*	*Plant F*	*Plant G*	*Plant H*	*Plant I*
				MICRO/ULTRA FILTRATION					
Pre-Treatment	Drum filter	Coag/Floc/ Clarification/ Chloramina-tion	Strainers	Screen	Screen	Coagulation / Flocculation / Disc filter/ UV/ Chlorination	Drum filter	Longitudinal screen/ Chloramina-tion	Rotating gravity screen/ Chloramin-ation
Membrane technology supplier	Pall/Asahi Kasei	Pall/Asahi Kasei	Pall/Asahi Kasei	Pall/Asahi Kasei	Siemens/ Memcor	GE/Zenon	Norit	GE/Zenon	Siemens/ Memcor
Configuration	HF	HF	HF	HF	HF	HF	CT	HF	HF
Membrane type	Pressurised	Pressurised	Pressurised	Pressurised	Pressurised	Immersed	Pressurised	Immersed	Immersed
Pore Size (µm)	0.1	0.2	0.1	0.1	0.04	0.02	0.03	0.02	0.04
Total membrane area (m^2)	1,700	85,400	3,200	160,000	-	5,016	304,640	15,600	730,000
Operating flux(LMH)	35	65	60	44	47	27.8	65	34	33
TMP range (bar)	0.1	0.3	0.26	0.24	0.8	0.33	0.38	0.4	0.25
				REVERSE OSMOSIS					
Membrane technology supplier	Koch	Toray	Hydarnautics	Hydranautics	Toray	DOW	Toray	DOW	Hydranautics
Total membrane area (m^2)	1,451	4,000	43,200	427,000	-	2,433	-	4,002	580,000
Elements/vessel	6	7	7	7	-	6	-	6	7
No. stages	2	3	2	2	2	2	-	2	3
Range of flux (LMH)	21	20	16.8	18	17	21	-	20	20.4
Recovery (%)	80	85	85	80	75	75	85	80	85
Rejection (%)	93	92	93	93	-	94	-	91	97

Table 2-4 Feedwater quality

Site:	*Plant A*	*Plant B*	*Plant C*	*Plant D*	*Plant E*	*Plant F*	*Plant G*	*Plant H*	*Plant I*	***Avg***	***SD***
TDS	724	1020	805	677	1100	900	1280	700	950	***906.2***	***203.3***
COD	49	60	30	115	45	43	43	55	54	***54.9***	***24.2***
TSS	14.6	8.8	1.3	10	14	7.2	20	3	6	***9.4***	***6.0***
Turbidity	4.5	4.5	5	12	6	1.5	12	1.5	1.8	***5.4***	***4.1***

Table 2-5 Flux, TMP and permeability

Site:	*Plant A*	*Plant B*	*Plant C*	*Plant D*	*Plant E*	*Plant F*	*Plant G*	*Plant H*	*Plant I*	***Avg***	***SD***
Operating flux (LMH)	35	65	60	44	47	28	65	40	33	**46.3**	**14.0**
Temperature (°C)	16	24	26	28	28	23	25	15	24	**23.2**	**4.7**
Temperature-corrected flux (LMH)	31	73	72	56	60	31	75	35	37	**49.2**	**18.0**
TMP (bar)	0.26	0.3	0.26	0.24	0.8	0.28	0.38	0.3	0.25	**0.3**	**0.2**
Permeability(lmh/bar)	120	244	276	232	74	109	198.3	115	149	**164.8**	**74.7**

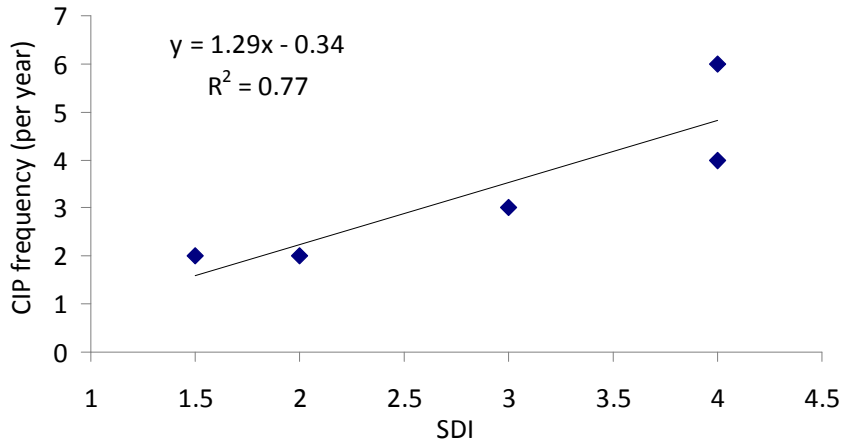


Figure 2-5 SDI impact on cleaning frequency of the RO membranes

Table 2-6 Membrane cleaning

Site:	*Plant A*	*Plant B*	*Plant C*	*Plant D*	*Plant E*	*Plant F*	*Plant G*	*Plant H*	*Plant I*
					MICRO/ULTRA FILTRATION				
Backwash frequency (min)	12	20	20 to 30	30	30	38	25	8 (wint) - 10 (summ)	22
Backwash water flux (m^3/h)	200	250	250	250-300	300	4, 3	250	100	170
Backwash air (m^3/h)						91	N/A	-	
CIP frequency	1-2/month	1/month	6/ year	1/month	1/month	1/month[(1)] 6/year[(2)]	1/year	1/month	21 days
CEB frequency per day/month	2/month	1/day [(1)] 1/week [(2)]	-	1-3/day	1/day	-	1/day	After 25-35 BW cycles	-
Chemical dosing for CIP	Alkaline NaOCl	NaOCl	Citric acid/NaOH	Citric acid	NaOCl	NaOCl[(1)] Citric acid[(2)]	Oxalic acid	NaOCl	NaOH+ Memclean/ citric acid
Chemical dosing for CEB	NaOCl	NaOCl[(1)] Citric Acid[(2)]	-	NaOCl/Citric acid	Citric acid	-	Cl_2 + NaOH	NaOCl	-
					REVERSE OSMOSIS				
Chemical dosing	Sulphuric	No	No	H_2SO_4	H_2SO_4	$NaHSO_3$	-	H_2SO_4	H_2SO_4
AntiScalant Dosing	Accepta 2651		Pretreat Plus100		-		-	-	AWC A-102 Plus
CIP frequency	6month	6 month	6 per year	-	-	6 per month	-	4-5/year	every 6 months
chemical dosing for CIP	Accepta 2068	HCl	citric acid	citric acid	-		-	NaOH	STPP

Table 2-7 Summary of RO Conversions for the sites

Site:	*Plant A*	*Plant B*	*Plant C*	*Plant D*	*Plant F*	*Plant H*	*Plant I*	***Avg***	***SD***
Product	Koch	Toray	UAT	Hydranautics	DOW	DOW	Hydranautics		
No. stages	2	3	2	2	2	2	3		
Elements per vessel	6	7	7	7	6	6	7		
Vessels/stage, Stage 1	4	120	72	64	8	20	78		
Vessels/stage, Stage 2	2	60	36	36	5	10	48		
Vessels/stage, Stage 3	na	30	na	na	0	na	24		
Overall Recovery	80	85	85	80	75	80	85	**81.4**	**3.8**
Per stage	55.3	46.87	61.2	55.3	50	55.3	46.9	**56.5**	**9.6**
Per element	12.55	8.64	12.67	10.86	12.6	12.6	8.64	**9.7**	**3.9**
Effluent TDS (mg/l)	52	80	58	45	56	60	30	**54.4**	**15.2**
TDS rejection(%)	93	92	93	93	94	91	97	**92.7**	**0.8**

2.3.2.4 Post-treatment

UV is used at 5 plants with 2 combining UV with hydrogen peroxide (advanced oxidation process, AOP). In wastewater reuse, UV and AOP are used both for organics removal - either by photolysis (UV) or both photolysis and hydrolysis (AOP) - and/or for disinfection (destruction of remaining microbial cells). However, disinfection by UV leaves no residual, demanding chemical disinfection by reagents such as chlorine (in three of the four plants surveyed). Since most of ions are removed by the RO process whilst the dissolved CO_2 passes into the permeate, the RO permeate is acidic and very low in TDS (Total dissolved solids). For some applications rehardening of the water and pH correction is required. The CO_2 may also be removed by stripping. When standards allow, RO permeate may be blended with MF/UF permeate to reduce or obviate further chemical addition, should the MF/UF permeate chemistry permit this.

2.3.3 Water quality

2.3.3.1 Feedwater quality

Key parameters in defining the operation and maintenance of the MF/UF process are the suspended solids, and in particular the turbidity which reflects the colloidal content, and organic carbon concentration (such as the chemical oxygen demand or COD). These parameters are normally interrelated to some extent since suspended/colloidal matter is largely organic (Table 2-4). Key parameters impacting on operation of the RO plant are the turbidity, total dissolved solids (TDS) and pH (Table 2-4). Both processes are influenced by temperature, which affects the viscosity and which demands that all flux and permeability values are corrected to a standard temperature of 20°C (Table 2-5). Temperature and pH were consistent across all sites, apart from the operating temperature at the Plants A and H sites which appear to be lower. Most sites appear to have high TDS levels, and consistent COD concentrations, apart from an anomalously high value for Plant D (also reported for Bedok,

Bartels *et al.*, 2007). Turbidity is also reasonably consistent with an average value of 17, but with higher values at Plant D and Plant G.

2.3.3.2 Microfiltration/ultrafiltration rejection

As expected, almost all the UF/MF membrane products generally achieve more than 97% removal of turbidity, and rather less COD exclusion (47% on average) which is presumably in the dissolved form (Table 2-8). There is some evidence to suggest that rejection of turbidity is related to membrane pore size, in that the 0.2 µm-rated membrane provides the highest fitrate turbidity and the 0.02 µm membrane the lowest (Figure 2-6). This is intuitive but rarely encountered in real plants since the membranes are normally protected by dynamic layer (Laitinen, 2002). On the other hand, given that the membranes are regularly backwashed this dynamic layer would be expected to be dislodged for some of the time. It is also noteworthy that the filtrate turbidity does not follow the feedwater turbidity. A high filtrate turbidity is of some practical consequence in that it may (a) exacerbate fouling of the RO membrane (Sadeddin et al, 2011), and (b) indicate the passage of viruses.

Table 2-8 MF/UF filtrate turbidity and COD concentration values

Product:	*Pall/Asahi kasei 1*	*Pall/Asahi kasei 2*	*Memcor*	*Norit*	*GE Zenon*	***Avg***	***SD***
Pore size	0.1	0.2	0.04	0.03	0.02		
Influent turbid, NTU	4.5	6	12	23.4	1.5	***9.48***	***8.67***
Effluent turbid, NTU	0.05	0.5	0.20	0.14	0.04	***0.186***	***0.19***
% removal, turbid.	***99***	***92***	***98***	***99***	***97***	***97.1***	***3.15***
Influent COD	40	85	38	30	56	***49.8***	***21.82***
Effluent COD	13	58	18	12	44	***29.0***	***20.81***
% removal, COD	***68***	***32***	***53***	***60***	***21***	***46.7***	***19.41***

2.3.3.3 Reverse osmosis rejection

The TDS rejection across all the sites is generally in the region of 97% for all but one installation (Table 2-3). This is lower than values achieved for seawater

desalination, where values >99.5% can be expected, since rejection is a function of the feed TDS concentration.

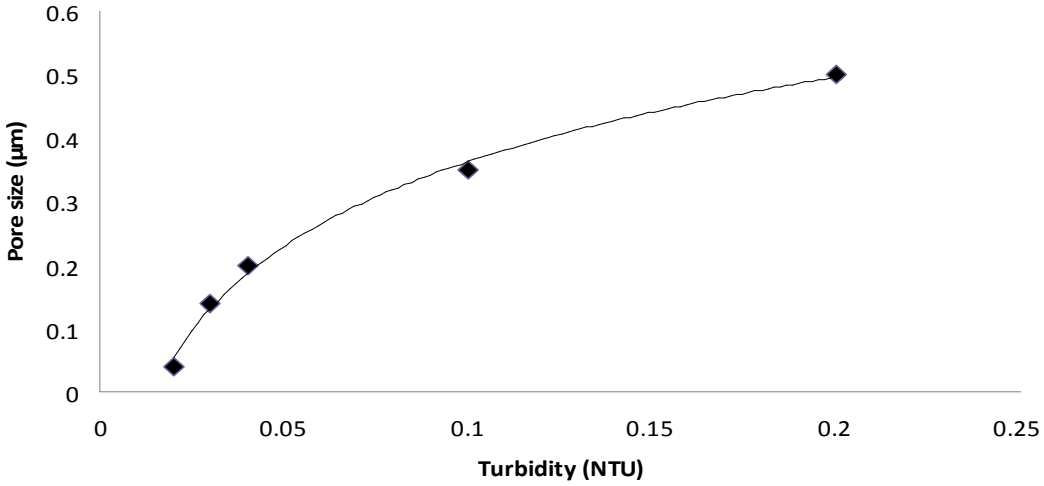


Figure 2-6 MF/UF pore size as a function of permeate turbidity

2.3.4 Specific energy demand

The overall specific energy demand (SED) is the energy consumption per unit permeate volume. For the membrane process components the SED varies unexpectedly widely from 0.8 to 2.3 kWh/m^3 across all sites, specific energy demand for pre- and post-treatment being negligible (Table 9). To assess this disparity, the SED for each process was compare with operating parameters (Figure 2-7 and Figure 2-8). Figure 2-7 clearly shows an exponential trend between the SED and flux. For the reverse osmosis, no clear trend was evident. However, it is known that an increase in flux or recovery necessarily increases the pressure – as does the TDS concentration in the case of RO - leading to increased energy consumption. RO SED increases with the product of these three parameters for all but one data point (Figure 2-8), though more data would be needed to verify this trend.

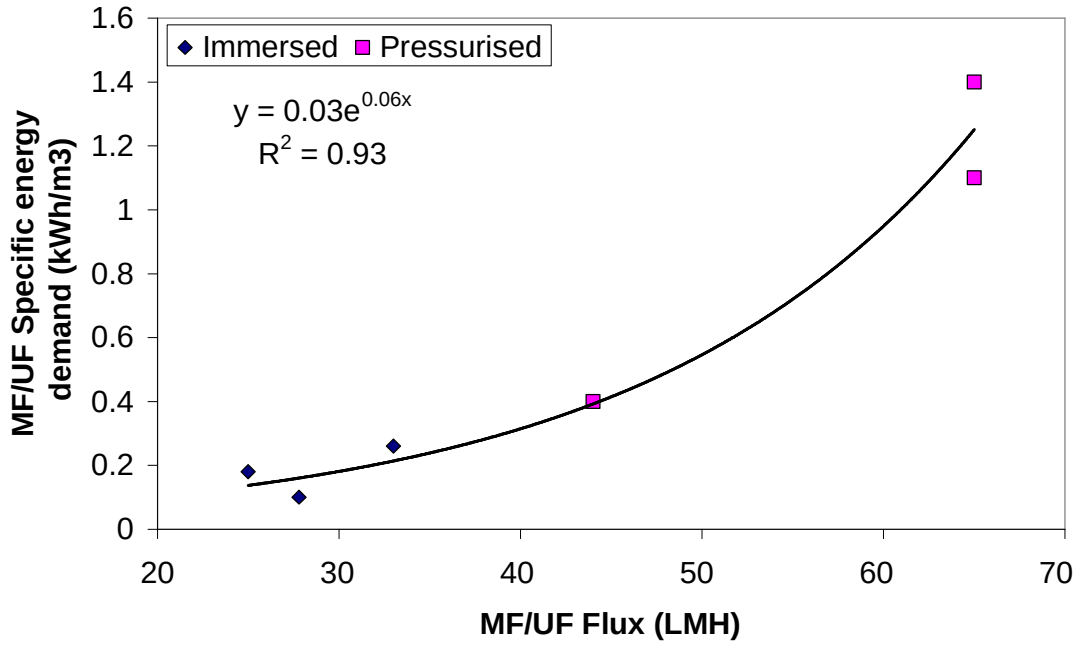


Figure 2-7 MF/UF specific energy demand as a function of the flux

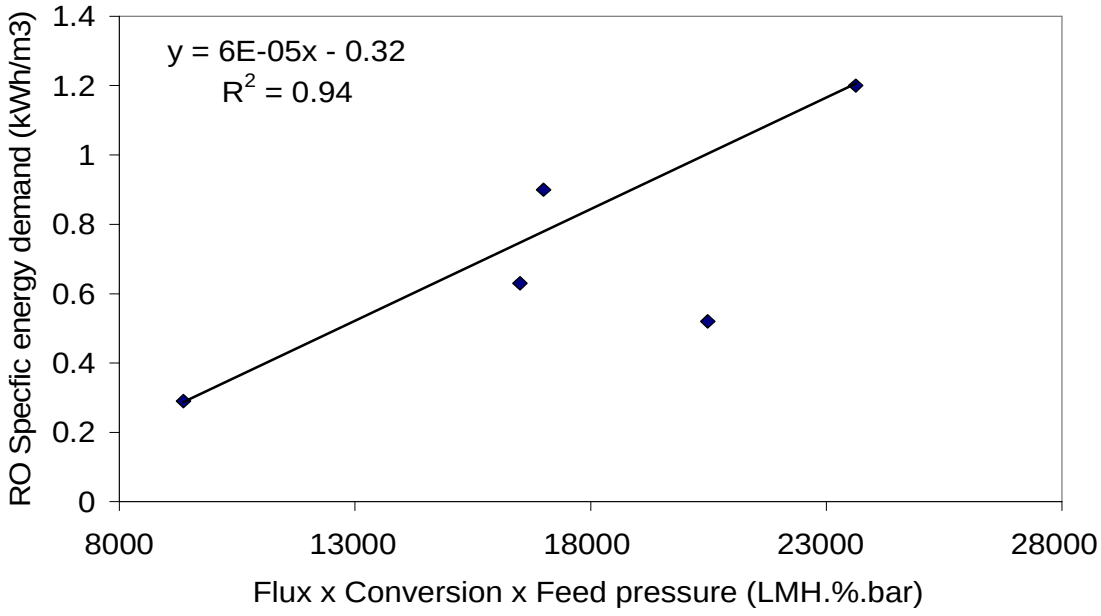


Figure 2-8 RO specific energy demand as a function of a coefficient being the multiplication of flux, recovery and feed pressure)

Table 2-9 Specific energy demand, kWh/m^3 permeate

Site:	*Plant A*	*Plant B*	*Plant D*	*Plant F*	*Plant G*	*Plant H*	*Plant I*
MF unit	-	1.4	0.4	0.1	1.1	0.18	0.26
RO unit	-	0.9	0.29	1.2	1.2	0.63	0.52
TOTAL	1-1.4	2.3	0.69	1.3	2.3	0.81	0.78

Whilst the energy demand values are higher than those associated with conventional wastewater treatment, generally 0.3-0.9 kWh/m^3 (Brepols *et al.*, 2008), they are also all significantly lower than the energy demand of seawater desalination which even for a state-of-the-art plant such as Ashkelon in Israel is >3.9 kWh/m^3 in total (Sauvet-Goichon, 2007). Such high energy demands for seawater desalination arise from feedwater TDS concentrations (35,000 – 50,000 mg/l) 40-60 times higher than those relating to the water reuse plants. The logic of water reuse using membrane technology as a sustainable alternative to seawater desalination is thus inescapable.

2.4 Conclusions

A review of nine membrane technology-based municipal wastewater reuse plants worldwide has revealed the following trends and plant facets:

- Pre-treatment of wastewater IMS plants is mostly through screening to remove large particles. Coagulation is used when TOC and phosphorus removal are needed. Chloramine may be added to suppress biofouling.
- Although backwash interval depends on the water quality and applied flux at each site, this operating parameter appears correlate reasonably well with feed water temperature. Chemical cleaning requirements are dependent mainly on the plant operating parameters and water quality.
- Microfiltration removes 97% of the turbidity and 47% of COD on average, with the membrane pore size for the MF/UF stage influencing the filtrate turbidity and thus the fouling of the downstream RO stage.
- Membrane permeability for the MF/UF stage is independent of the configuration of both the membrane and the module.

- The silt density index (SDI) of the feed to the RO stage impacts on the chemical cleaning requirement, with cleaning frequency increasing with SDI.
- The reported specific energy demand values range from 0.8 to 2.3 kWh/m^3 permeate, or around 25-70% of the energy demand for seawater desalination. MF/UF specific energy demand was dependent on flux while the RO specific energy demand was apparently dependent on the product of the flux, recovery and feed pressure.

2.5 References

Asano, T. (2006), *Water reuse: issues, technologies, and applications*, Metcalf and Eddy, 1st Ed., McGrawHill, New York.

Bartels, C., Wilf, M., Andes, K., Long, J. (2007), Design considerations for wastewater treatment by reverse osmosis, *Water and Wastewater Asia*, May/June 2007, p. 20-26.

Brepols, C. (2011), *Operating large scale membrane bioreactors for municipal wastewater treatment*, IWA Publishing, 1st Ed., London, UK

Cazurra, T. (2008), Water reuse of south Barcelona's wastewater reclamation plant, *Desalination*, 218, p. 43-51.

Citulski, J.A., Farahbakhsh, K., Kent, F.C. (2009), Effects of total suspended solids loading on short-term fouling in the treatment of secondary effluent by an immersed ultrafiltration pilot system, *Water Environment Research*, 81, p. 2427-2436.

Du Pisani, P. L. (2006), Direct reclamation of potable water at Windhoek's Goreangab reclamation plant, *Desalination,* 188 (1-3, p. 79-88.

Esteban R. I., Ortega de Miguel, E. (2008), Present and future of wastewater reuse in Spain, *Desalination*, 218, p. 105-119.

Freeman, S., Bates, J., Wallis-Lage, C., McEvoy, J. (2008), Drought relief in South East, Queensland, Australia, provided by membrane-reclaimed water, *AWWA Journal*, 100 (2), p. 40-52.

Gabelich, C. J., Ishida, K.P., Gerringer, W., Evangelista, R., Kalyan, M., Suffet, I.H. (2006), Control of residual aluminium from conventional treatment to improve reverse osmosis performance, *Desalination*, 190, p. 147-160.

Gómez Gotor, A., Pérez Baez, S.O., Espinoza, C.A., Bachir, S.I. (2001), Membrane processes for the recovery and reuse of wastewater in agriculture, *Desalination*, 137, p. 187-192.

Greenberg, G., Hasson, D., and Semiat, R. (2005), Limits of RO recovery imposed by calcium phosphate precipitation, *Desalination*, 183, p. 273-288.

Hatt, J., Germain, E., and Judd, S.J. (2011), Screening optimisation for indirect potable reuse, *Water Science and Technology*, 63 (12), p. 2846-2852.

Jimenez, B., Asano T. (2008), *Water Reuse, an international survey of current practice, issues and needs*, 1st Ed., IWA Publishing, London, UK.

Judd, S. (2010), *The MBR book: principles and applications of membrane bioreactors in water and wastewater treatment*, 2nd Ed., Elsevier, Oxford, UK.

Kimura, K., Yamamura, H. (2006), Irreversible fouling in MF/UF membranes caused by natural organic matters (NOMs) isolated from different origins, *Separation Science and Technology*, 41, p. 1331-1344

Latinen N. (2002), Development of a ceramic membrane filtration equipment and its applicability for different wastewaters, PhD thesis, Lappeenranta University of Technology.

Lazarova, V., Shields, P., Levine, B., Savoye, P., Hranisavljevic, D., Renaud, P. 92003), Production of high quality for reuse purposes: the West Basin experience, *Water Science and Technology: Water Supply*, 3 (3), p. 167-175.

Liu, Q-F., Kim, S-H., Lee, S. (2009), Prediction of microfiltration membrane fouling using artificial neural network models, *Separation and Purification Technology*, 70, p. 96-102.

MacBeth, I., Murrer, J., Latter, S. (2004), Reducing the costs for ultra-pure water production for use as de-min plant feed water for Peterborough power station, UK, Chemie im Kraftwerk 2004.

Markus, M.R., Deshmukh, S.S. (2010), An innovative approach to water supply - The Groundwater Replenishment System, In: *World Environmental and Water Resources Congress 2010: Challenges of Change*, Providence, Rhode Island 16-20 May 2010, p. 3624-3639.

Moon, J., Kang, M-S., Lim, J-L., Kim, C-H., Park, H-D. (2009), Evaluation of a low-pressure membrane filtration for drinking water treatment: pre-treatment by coagulation/sedimentation for the MF membrane, *Desalination*, 247(1-3), p. 271-284.

Psoch, C., Shiewer, S. (2006), Resistance analysis for enhanced wastewater membrane filtration, *Journal of Membrane Science*, 280, p. 284-297.

Soice, N.P., Maladono, A.C., Takigawa, D.Y., Norman, A.D., Krantz, W.B., Greenberg, A.R. (2003), Oxidative degradation of polyamide RO membranes: studies of molecular model compounds and selected membranes, *Journal of Applied Polymer Science*, 90 (5), p. 1173-1184.

Rodriguez, D., Van Buynder, P., Lugg, R., Blair, P., Devine, B., Cook, A. and Weinstein, P. (2009), Indirect potable reuse: A sustainable water supply alternative, *International Journal of Environmental Research and Public Health,* 6 (3), p. 1174-1209.

Sadeddin, K., Naser, A., Firas, A. (2011), Removal of turbidity and suspended solids by electro-coagulation to improve feed water quality of reverse osmosis plant, *Desalination*, 268, p. 204-207.

Sauvet-Goichon B. (2007), Ashkelton desalination plant – a successful challenge, *Desalination*, 203, p. 75-81.

Wilf, M. (2010), *The guidebook to membrane technology for wastewater reclamation*, 1st Ed., Balaban Desalination Publications, Hopkinton, USA.

Van Houtte, E., Verbauwhede, J. (2008) Operational experience with indirect potable reuse at the Flemish Coast, *Desalination*, 218, p. 198–207.

Wang, L., Wang, X., Fukushi, K-I. (2008), Effects of operational conditions on ultrafiltration membrane fouling, *Desalination*, 229, p. 181-191.

Xu, P., Bellona, C., Drewes, L.E. (2010), Fouling of nanofiltration and reverse osmosis membranes during municipal wastewater reclamation: Membrane autopsy results from pilot-scale investigations, *Journal of Membrane Science*, 353, p. 111-121.

CHAPTER 3: STATISTICAL EXPERIMENTAL PROGRAMMING FOR MEMBRANE PROCESS OPTIMISATION

Daviu, R., Raffin, M., Germain, E., Judd, S., Statistical experimental programming for membrane process optimisation, submitted to *Membranes.*

3.1 Indirect potable reuse process optimisation

The recovery of municipal wastewater for indirect potable reuse (IPR) is viewed as a viable option for conserving freshwater resources in many arid regions of the world (Rodriguez *et al.*, 2009). The technical feasibility of using membrane technology for thus duty, in order to provide a reliable and high product water quality which is robust to changes in feedwater quality, has been demonstrated by several studies (Markus and Deshmukh, 2010) and a number of full-scale installations (Chapter 2 and Annex 1). Currently, dozens of IPR schemes exist worldwide principally located in US (e.g.: groundwater replenishment system, Orange county, Markus and Deshmukh, 2010), Singapore (NeWater plants, Seah *et al.*, 2008), Australia (e.g.: Western Corridor recycled water project, Roux *et al.*, 2010), and Europe (Torreele – IWVA, Belgium, Van Houtte and Verbauwhede, 2008). Only one scheme, in Namibia, is dedicated to direct potable reuse (Windhoek's Goreangab reclamation plant, Du Pisani, 2006)), i.e. where the water is used without passing through an environmental water body. Most of these schemes employ a multi-barrier approach to remove contaminants, which includes microfiltration (MF) or ultrafiltration (UF) membranes, reverse osmosis (RO) membranes and in some cases UV irradiation for disinfection.

Optimisation of IPR membrane systems at pilot scale is usually through classical correlations of a single impacted parameter (such as flux or permeability) against a single variable (such as chemical reagent concentration or cleaning intensity). Normally such optimisation relates to minimisation of membrane fouling, since this is ubiquitous in membrane technology. However, such a classical approach necessarily limits the number of parameters that may be rigorously studied, such that pilot plant studies rarely encompass all combinations of all key variables in studying impacts on fouling.

Recent studies in membrane technology and process development have made use of Box-Behnken experimental programme design for process optimisation,

including the influence of operating parameters (Lin *et al*., 2008) or membrane material (Sivakumar *et al.*, 1999) on UF membrane rejection and also on fouling minimisation (Jokic *et al.*, 2010) or on chemical cleaning (Porcelli *et al.*, 2010a, 2010b). Optimisation implies identification of the most effective operating conditions for control of fouling, and scaling in particular in the case of reverse osmosis since this is the main limitation of dense membrane processes (Daramola *et al.*, 2007; Hosam-Eldin *et al.*, 2009). It is thus of interest to establish the relative efficacy of all statistical experimental programming techniques. This paper compares five statistical methods with respect to the number of experiments, the simplicity of calculation, the order of the response, the estimation of error and the distribution of information throughout the range of parameter values of interest, which are among the 14 properties identified as contributing to optimum experimental design (Box and Draper, 1987).

3.2 Literature review

3.2.1 Membrane optimisation and statistical design

Whilst the conventional method of optimisation is through adjusting a single variable at a time while maintaining constant other parameter values, this method demands a large number of experiments and so consumes extensive periods of time and incurs high costs. Moreover, synergistic effects of the various parameters studied are not necessarily accounted for such that true optimisation cannot be ensured (Khayet *et al.*, 2008). Statistical methods for experimental programme design can instead limit the number of tests required to identify the optimum condition (Porcelli and Judd, 2010; Jokić *et al.*, 2010), reducing the time and costs.

Experimental planning may be based on either a first or a second order model. A first order model provides a first approximation distinguishing between those factors having a significant impact on a process and those which do not. This can be produced by applying a 2-level design, and can be used when a high

number of parameters need to be screened to study their effect in the response. A more complex method with the identified parameters affecting the process response may then be applied (Tarley *et al.*, 2009). Furthermore, many processes cannot be represented by a first order model and require second order models. For this, a three or higher level design is required.

3.2.2 Statistic methods

Available literature indicates five main statistical methods employed for process optimisation: factorial design (2 and 3 level), Taguchi, Central Composite Design (CCD), Doehlert and Box-Behnken Design (BBD). Examples of papers published in learned journals relating specifically to membrane separation technology for various applications are summarised in Figure 3-1. This figure clearly indicates factorial design to be the most commonly used, with the BBD being amongst the least popular with only nine publications found. These methods have been applied to a wide range of processes (Table 3-1).

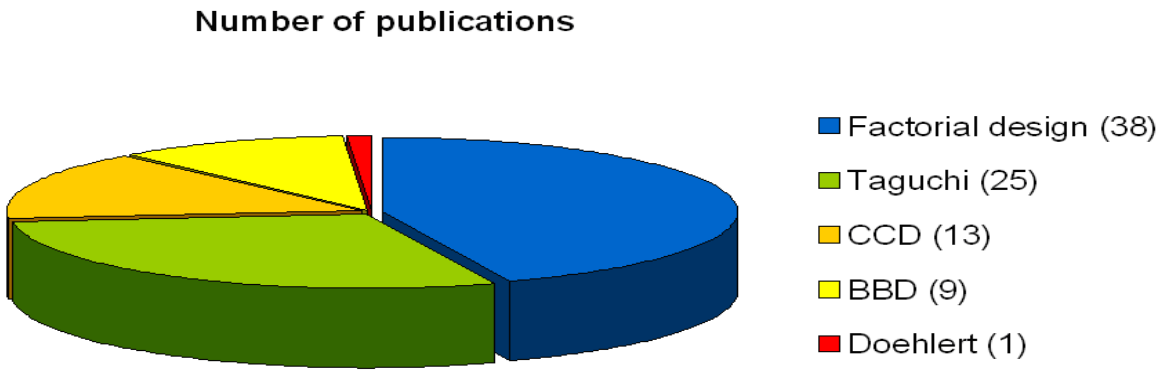


Figure 3-1 Number of published studies employing statistical methods in membrane separation technology research across various applications.

3.2.3 BBD vs. other methods

BBD may be defined as a three-level fractional factorial design which does not take into account the combinations in which all the parameters are at their lowest or highest levels. Many studies advocate the use of BBD specifically for

process optimisation (Ferreira *et al.*, 2007; Hinkelmann and Jo, 1998). A review of the literature (*Scopus* search: “Box” and “Behnken”) reveals BBD to have been employed in 1143 studies across a wide range of disciplines including chemistry, microbiology, biochemistry, pharmaceuticals, food, water and wastewater and materials (Figure 3-2). 99 studies have been published in the In water and wastewater areas (Figure 3-3), four relating to membranes and 70% to wastewater. The progression of BBD as applied to water and wastewater treatment reveals an increasing trend with only three studies reported in 2000 compared with 27 and 26 in 2009 and 2010 respectively (Figure 3-4).

Table 3-1 Examples of application of the statistical methods

Method	*Application*	*Ref*
2^k	• Study of influence of operating parameters in a juice concentration process using membrane and osmotic distillation.	Onsekisoglu *et al.* (2010)
	• Process modelling of hydraulic backwash of membranes.	Daramola *et al.* (2007)
3^k	• Investigation of the effects of feed temperature, sludge retention time and organic loading rate for a membrane bioreactor	Birima *et al.* (2009)
	• Membrane characterization by designing a thin channel cross-flow	Darcovich *et al.* (1997)
Taguchi	• Investigation of the permeate flux of wastewater in function of the operation conditions	Hesampour *et al.* (2008)
	• Optimisation of the cleaning in place for a nanofiltration system	Gönder *et al.* (2010)
CCD	• Optimisation of a coagulation-flocculation hybrid process prior to membranes in drinking water.	Zularisam *et al.* (2009)
	• Separation process optimisation of p-xilene from p-/o-xylene through a membrane, studying the effect of the temperature, the partial pressure of the p-xilene and its feed composition	Yeong *et al.* (2009)
BBD	• Optimisation of operating parameters for microfiltration of Baker’yeast suspension	Jokić *et al.* (2010)
	• Investigation of the effect of water, methanol, soap and glycerol in the purification of biodiesel by employing membrane technology	Saleh *et al.* (2010)
Doehlert	• Determination of dinitrophenolic trace levels in water for	Bartolomé *et al.* (2007)

	a membrane system

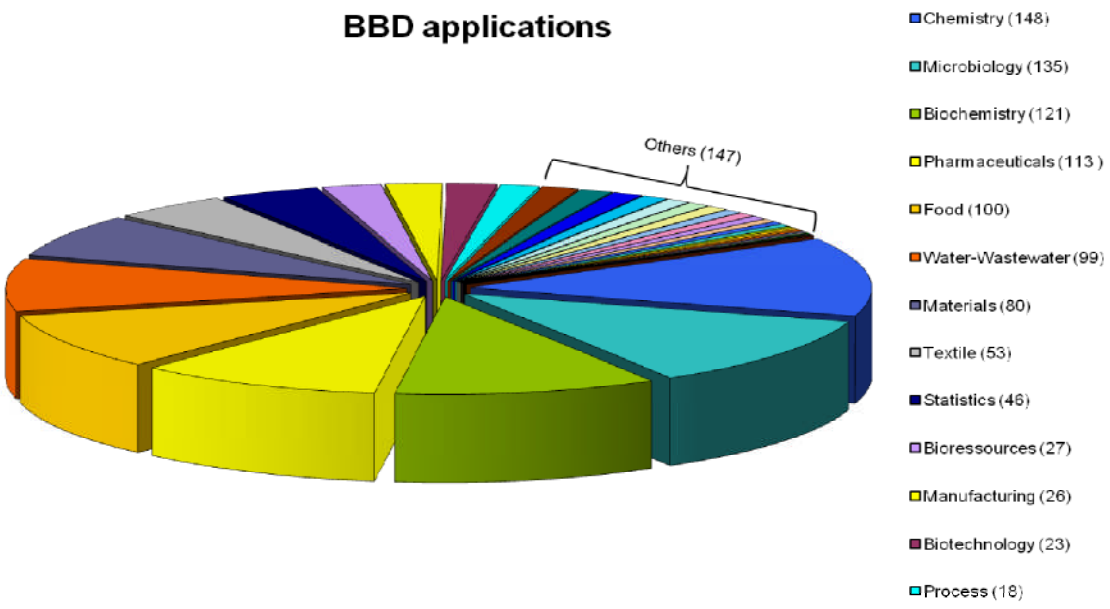


Figure 3-2 Number of BBD studies for different applications

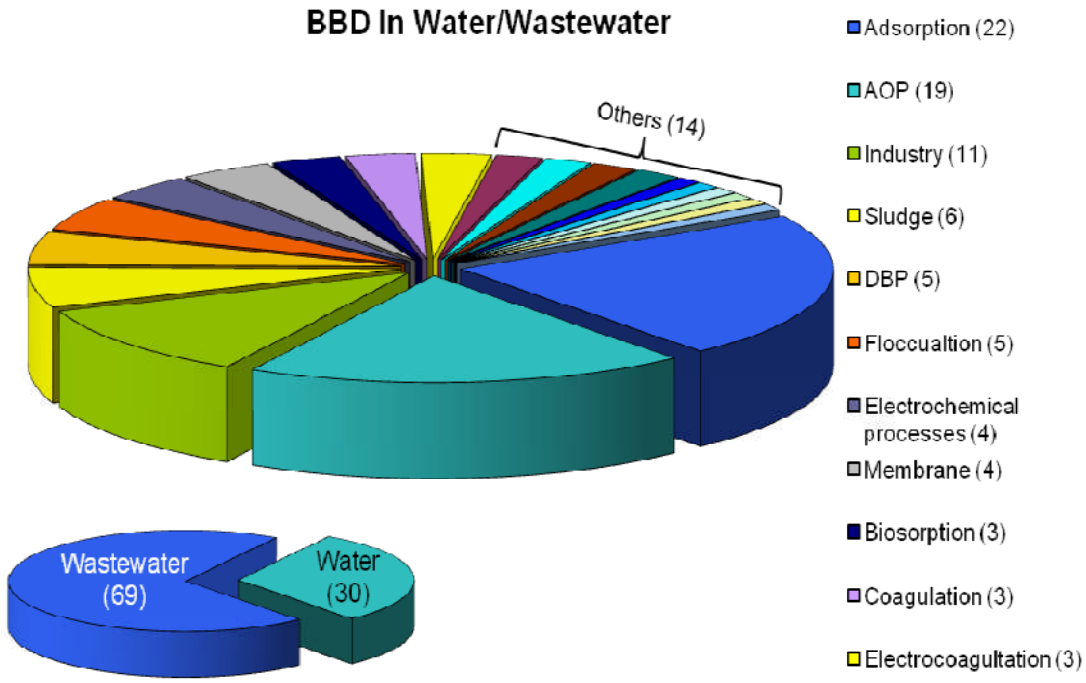


Figure 3-3 Number of BBD studies for different applications in water and wastewater

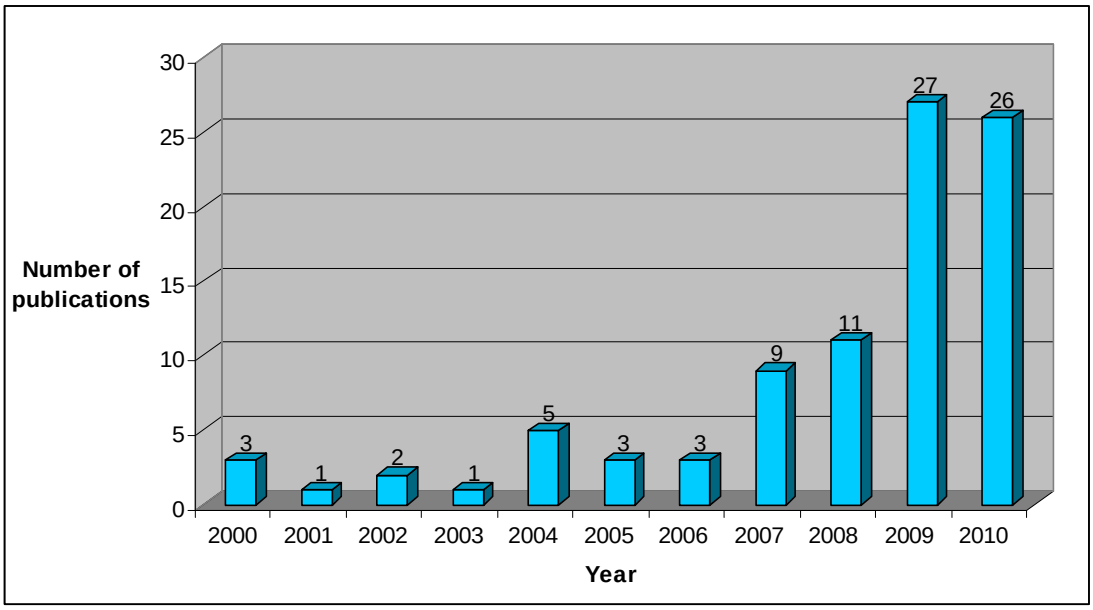


Figure 3-4 Number of BBD studies published per year

A summary of the methods and their facets (Table 3-2) reveals them to differ significantly with respect to the required number of experiments and the order of the response provided (first order or higher), and so the overall efficiency of the method. The number of experiments depends on the number of parameters studied and the level, which means the number of values applied to study for each parameter. Figure 3-5 shows the number of tests required to analyse 2-5 parameters, all at three levels (the minimum required to generate a quadratic response and so assess the non-linearity of a system). The number of runs needed includes three replicates for the central point in the required cases. For example, for a study with only two parameters, BBD is the method demanding the least number of experiments (7) and CCD the one needing the most (11). In the case of three parameters the Taguchi experimental design requires the least number of tests (9) followed by BBD and Doehlert, (both requiring 15). When four parameters are considered Taguchi requires fewer experiments, and BBD and CCD require three times less experiments than the 3-level design (27 and 81 respectively). Thus, the order of preference for the different methods in

terms of the required number of experiments changes according to the number of parameters to be studied in the tests:

- Two parameters: BBD<Taguchi, Doehlert, 3-level<CCD
- Three parameters: Taguchi<BBD, Doehlert<CCD<3-level
- Four parameters: Taguchi<Doehlert<BBD, CCD<3-level
- Five parameters: Taguchi<Doehlert<CCD<BBD<3-level

A further important aspect to be considered in selection of an experimental design is the order of the equation generated, since this defines whether the relationship is linear or non-linear. A first order model provides a linear response while a second order model generates a quadratic response and allows synergistic effects to be assessed through determination of the interaction coefficient. As mentioned already, the 2-level factorial design is not able to produce second order or higher order model and so cannot be used to optimise processes with quadratic responses. This also applies to Taguchi when only 2 levels are considered.

Table 3-2 Summary of the main points and advantages/disadvantages of the experimental plans

Method	***Nº of exp.***	***Advantages/ Disadvantages***
2-level Factorial	2^n	+Simple to apply, relatively low cost and very useful as a first screening (Tarley *et al.*, 2009) -Not valid for quadratic responses
3-level Factorial	3^n	+Simple to apply, relatively low cost and very useful as a first screening (Tarley *et al.*, 2009) -Large number of experiments is required for more than 2 parameters
Taguchi	2 level: 4 (2, 3 param.); 8 (4, 5 param.) 3 level: 9 (2, 3, 4 param.); 18 (5 param.)	+Low number of experiments required -Not efficient for interactions (Nair *et al.*, 1992).
CCD	$N=k^2+2k+C_p$	+All factors are studied in five levels -The five levels studied are not equidistant
BBD	$N=2k(k-1)+C_p$	+ Equidistant levels for the variables +/-Factors at their highest or lowest level are not considered at the same time, which is an advantage or disadvantage depending on the process
Doehlert	$N=k^2+k+C_p$	+Uniformity in the experimental domain for the variables -A first screening is recommended to know the most significant factor to apply at the highest level

K, n: number of parameters, N:number of expedients needed, Cp: number of repetitions at central point

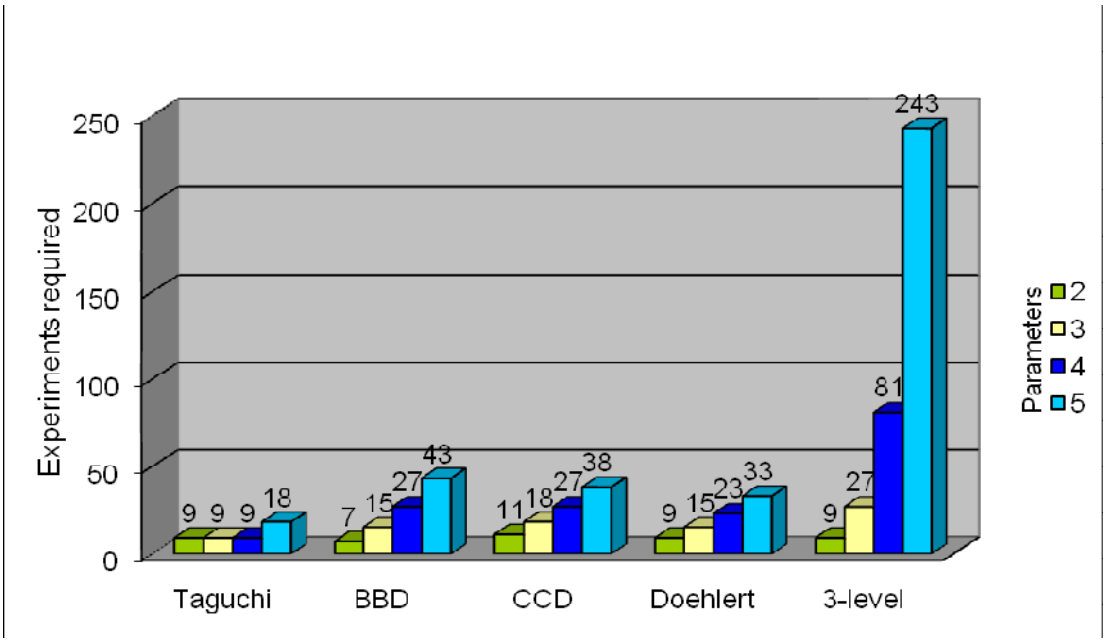


Figure 3-5 Number of experiments required by each method for 2, 3, 4 and 5 parameters

Factorial design (full or fractional) has been widely used for generating process models in water-related subjects (Onsekizoglu *et al.*, 2010; Yi *et al.*, 2011; Wu *et al.*, 2008; Mosteo *et al.*, 2006). A full factorial design may be a more appropriate tool for complex processes for quantifying the interactions amongst the variables under study (Rajasimman *et al.*, 2009). 2-level factorial design is the most commonly used among the first order models, since this is simple and generally economical to apply (Tarley *et al.*, 2009). This design I also used to screen an excessive number of variables so as to reduce their number to permit a more rigorous experimental design. 3-level factorial design is used for more complex systems where the process cannot be represented by a first order model, i.e. when a second order model is required to generate a non-linear response. However, this then requires a large number of experiments, which increases with the number of parameters *n* studied (according to 3^n). For this reason, its application in response surface modelling (which is a collection of

statistical and mathematical techniques for the determination of an empirical model (Khayet *et al.*, 2008) is limited when the number of factors is higher than two (Bezerra *et al.*, 2008).

Central composite design is considered as a better alternative to the 3-level full factorial design since it requires less experimentation whilst producing similar results (Tarley *et al.*, 2009). The main disadvantage of this method is that the studied levels of the parameters are not equidistant (5-level design: –α,-1,0,+1,+α), such that there is the possibility of the optimum value arising between the largest gap between the analysed values.

In the case of the Taguchi method the aim is to identify conditions with minimum sensitivity to noise, which comprises all the external variables that are not controllable but nonetheless affect the process. This provides a robust method for experimental design (Besseris, 2008), and the method been widely applied in fields including microbiology, agriculture, chemistry and engineering, but not so much for membrane technology (Gönder *et al.*, 2010). Although Taguchi is often used, since it requires the least number of experiments, there is a long-standing debate as to the efficiency of this method to assess the interaction between the parameters analysed (Nair *et al.*, 1992). It may therefore be inappropriate for processes where synergistic effects are significant.

The Doehlert method is recommended as an experimental plan in reviews of statistical methods for process optimisation (Tarley *et al.*, 2009; Bezerra *et al.*, 2008) though it is preferable to know the most significant factor for the process since this should be associated with the highest level (Tarley *et al.*, 2009). This then demands either a wide knowledge of the process or else a screening trial to identify the most significant parameter, which then necessarily increases the required number of experiments.

According to Ferreira *et al.* (2007), BBD and Doehlert matrix are slightly more efficient than central composite design but much more efficient than the three-level factorial designs in terms of ratio of the number of coefficients in the generated model algorithm and the number of experiments required to produce

it. In applying BBD to membrane optimisation studies specifically, its effectiveness in detecting interactions between parameters that are undetected using conventional methods becomes advantageous. Such interactions between parameters pertaining to membrane process optimisation have been reported from previous studies using BBD (Porcelli *et al.*, 2010a, 2010b; Figueroa *et al.*, 2011). Moreover this is one of the methods requiring the fewest number of experiments for equidistant parameter values, providing a more uniform investigation. The method does not demand experiments employing the variables set at their highest and/or lowest values, thereby avoiding extreme conditions which may be inappropriate for the sustainable functioning of the membranes.

3.3 Conclusions

The use of statistical experimental programming methods in water and wastewater treatment in general, and membrane processing specifically, has been reviewed. Five methods are identified from publications in the peer reviewed literature, the most popular being factorial design. Statistical methods were compared with respect to the number of experiments.

It is concluded that Box-Behnken offers an appropriate and efficient method for experimental design to optimise membrane processes and has been successfully demonstrated in a multitude of scientific fields. The method has increased in popularity but remains marginal in its application, with only 10% of total publications in the area in the peer reviewed-literature within the past 10 years employing this method. BBD requires a smaller number of experiments than that of 3-level factorial design, decreasing costs and required experimental time. Other methods such as Taguchi or Doehlert design require fewer experiments than BBD when the number of parameters increases beyond 3.

Since BBD provides a 3-level design, it allows 2^{nd} order algorithms to be defined which can then describe non-linearity of a system and the interaction between parameters; this is not possible with a 2-level design. BBD permits simple replication of the identified optimised conditions (defined by the “central point”)

allowing precision of the output to be determined as well as errors arising from the practical experimentation method. This central point is also used with CDD and Doehlert. Contrary to CDD and Doehlert, levels are equidistant, which then provides uniform distribution of the parameter values within the experimental region of interest.

Finally, BBD does not demand experiments employing the variables set at extreme values which may otherwise adversely affect the system, such as permanent membrane fouling.

3.4 References

Bartolomé, L., Lezamiz, J., Etxebarria, N., Zuloaga, O. and Jönsson, J. A. (2007), Determination of trace levels of dinitrophenolic compounds by microporous membrane liquid-liquid extraction in environmental water samples, *Journal of Separation Science,* 30 (13), p. 2144-2152.

Besseris, G. J. (2008), Multi-response optimisation using Taguchi method and super ranking concept, *Journal of Manufacturing Technology Management,* 9 (8), p. 1015-1029.

Bezerra, M. A., Santelli, R. E., Oliveira, E. P., Villar, L. S. and Escaleira, L. A. (2008), Response surface methodology (RSM) as a tool for optimization in analytical chemistry, *Talanta,* 76 (5), p. 965-977.

Birima, A. H., Mohd Noor, M. J. M., Mohammed, T. A., Idris, A., Muyibi, S. A., Nagaoka, H., Ahmed, J. and Ghani, L. A. A. (2009), The effects of SRT, OLR and feed temperature on the performance of membrane bioreactor treating high strength municipal wastewater, *Desalination and Water Treatment,* 7 (1-3), p. 275-284.

Box, G.P., Drapaer, N.R. (1987), *Empirical Model-Building and Response surfaces*, John Wily and Sons, Inc., USA, p 477

Daramola, M. O., Keesman, K. J. and Spenkelink, F. (2007), Process modelling of ultrafiltration units: An RSM approach, *Journal of Applied Sciences,* 7 (23), p. 3687-3695.

Darcovich, K., Dal-Cin, M. M., Ballèvre, S. and Wavelet, J. (1997), CFD-assisted thin channel membrane characterization module design, *Journal of Membrane Science,* 124 (2), p. 181-193.

Du Pisani, P. L. (2006), Direct reclamation of potable water at Windhoek's Goreangab reclamation plant, *Desalination,* 188 (1-3), p. 79-88.

Ferreira, S. L. C., Bruns, R. E., Ferreira, H. S., Matos, G. D., David, J. M., Brandão, G. C., da Silva, E. G. P., Portugal, L. A., dos Reis, P. S., Souza, A. S. and dos Santos, W. N. L. (2007), Box-Behnken design: An alternative for the optimization of analytical methods, *Analytica Chimica Acta,* 597 (2), p. 179-186.

Figueroa, R. A., Cassano, A. and Drioli, E. (2011), Ultrafiltration of orange press liquor: Optimization for permeate flux and fouling index by response surface methodology, *Separation and Purification Technology,* 80 (1), p. 1-10.

Gönder, Z. B., Kaya, Y., Vergili, I. and Barlas, H. (2010), Optimization of filtration conditions for CIP wastewater treatment by nanofiltration process using Taguchi approach, *Separation and Purification Technology,* 70 (3), p. 265-273.

Hesampour, M., Krzyzaniak, A. and Nyström, M. (2008), The influence of different factors on the stability and ultrafiltration of emulsified oil in water, *Journal of Membrane Science,* 325 (1), p. 199-208.

Hinkelmann, K. and Jo, J. (1998), Linear trend-free Box-Behnken designs, *Journal of Statistical Planning and Inference,* 72 (1-2), p. 347-354.

Hossam-Eldin, A., El-Nashar, A. M. and Ismaiel, A. (2009), Techno-economic optimization of SWRO desalination using advanced control approaches, *Desalination and Water Treatment,* 12 (1-3), p. 389-399.

Jokić, A., Zavargo, Z., Šereš, Z. and Tekić, M. (2010), The effect of turbulence promoter on cross-flow microfiltration of yeast suspensions: A response surface methodology approach, *Journal of Membrane Science,* 350 (1-2), p. 269-278.

Khayet, M., Cojocaru, C. and Zakrzewska-Trznadel, G. (2008), Response surface modelling and optimization in pervaporation, *Journal of Membrane Science,* 321(2), p. 272-283.

Lin, S-H., Hung, C-L., Juang R-S. (2008), Effect of operating parameters on the separation of proteins in aqueous solutions by dead-end ultrafiltration, *Desalination*, 234, p. 116-125

Markus, M.R., Deshmukh, S.S. (2010), An innivative approach to water supply - The Groundwater Replenishment System, In: *World Environmental and Water Resources Congress 2010: Challenges of Change*, Providence, Rhode Island 16-20 May 2010, p. 3624-3639.

Mosteo, R., Ormad, P., Mozas, E., Sarasa, J. and Ovelleiro, J. L. (2006), Factorial experimental design of winery wastewaters treatment by heterogeneous photo-Fenton process, *Water research,* 40 (8), p. 1561-1568.

Nair, V. N., Abraham, B., MacKay, J., Nelder, J. A., Box, G., Phadke, M. S., Kacker, R. S., Sacks, J., Welch, W. J., Lorenzen, T. J., Shoemaker, A. C., Tsui, K. L., Lucas, J. M., Taguchi, S., Myers, R. H., Vining, G. G. and Wu, C. F. J. (1992), Taguchi´s Parameter Design: A panel discussion, *Technometrics,* 34 (2), p. 127-161.

Onsekizoglu, P., Savas Bahceci, K. and Acar, J. (2010), The use of factorial design for modeling membrane distillation, *Journal of Membrane Science,* 349 (1-2), p. 225-230.

Porcelli, N. and Judd, S. (2010a), Chemical cleaning of potable water membranes: The cost benefit of optimisation, *Water research,* 44 (5), p. 1389-1398.

Porcelli, N. and Judd, S. (2010b), Impact of cleaning protocol on membrane permeability recovery: a sensitivity analysis, *J. AWWAF*, 102 (12)

Rajasimman, M., Sangeetha, R. and Karthik, P. (2009), Statistical optimization of process parameters for the extraction of chromium(VI) from pharmaceutical wastewater by emulsion liquid membrane, *Chemical Engineering Journal,* 150 (2-3), p. 275-279.

Rodriguez, D., Van Buynder, P., Lugg, R., Blair, P., Devine, B., Cook, A. and Weinstein, P. (2009), Indirect potable reuse: A sustainable water supply alternative, *International Journal of Environmental Research and Public Health,* 6 (3), p. 1174-1209.

Roux, A., Robillot, C., Chapman, H., Leusch, F., Hodge, M., Walker, T. (2010), Hazard identification, qualitative risk assessment and monitoring on the Western Corridor Recycled Water Project, In: *7th IWA Specialty Conference on Wastewater Reclamation & Reuse, 21-25 September 2009, Brisbane, Australia*.

Saleh, J., Dubé, M. A. and Tremblay, A. Y. (2010), Effect of soap, methanol, and water on glycerol particle size in biodiesel purification, *Energy and Fuels,* 24 (11), p. 6179-6186.

Seah, H., Tan, T.P., Chong, M.L. and Leong, J., (2008), NEWater-multi safety barrier approach for indirect potable use. Water Science & Technology: Water Supply, 8 (5), p. 573–588.

Sivakumar, M., Annadurai, G., Mohan, D. (1999), Studies on Box-Behnken design experiments : cellulose acetate-polyurethane ultrafiltration membranes for BSA separation, *Bioprocess Engineering*, 21, p. 65-68.

Tarley, C. R. T., Silveira, G., dos Santos, W. N. L., Matos, G. D., da Silva, E. G. P., Bezerra, M. A., Miró, M. and Ferreira, S. L. C. (2009), Chemometric tools in electroanalytical chemistry: Methods for optimization based on factorial design and response surface methodology, *Microchemical Journal,* 92 (1), p. 58-67.

Van Houtte, E. and Verbauwhede, J. (2008), Operational experience with indirect potable reuse at the Flemish Coast, *Desalination,* 218 (1-3), p. 198-207.

Wu, J., Le-Clech, P., Stuetz, R. M., Fane, A. G. and Chen, V. (2008), Novel filtration mode for fouling limitation in membrane bioreactors, *Water research,* 42 (14), p. 3677-3684.

Yeong, Y. F., Abdullah, A. Z., Ahmad, A. L. and Bhatia, S. (2009), Process optimization studies of p-xylene separation from binary xylene mixture over silicalite-1 membrane using response surface methodology, *Journal of Membrane Science,* 341 (1-2), p. 96-108.

Yi, X. S., Shi, W. X., Yu, S. L., Li, X. H., Sun, N. and He, C. (2011), Factorial design applied to flux decline of anionic polyacrylamide removal from water by modified polyvinylidene fluoride ultrafiltration membranes, *Desalination,* 274 (1-3), p. 7-12.

Zularisam, A. W., Ismail, A. F., Salim, M. R., Sakinah, M. and Matsuura, T. (2009), Application of coagulation-ultrafiltration hybrid process for drinking water treatment: Optimization of operating conditions using experimental design, *Separation and Purification Technology,* 65 (2,) p. 193-2.

CHAPTER 4: OPTIMISING OPERATION OF AN INTEGRATED MEMBRANE SYSTEM (IMS) – A BOX-BEHNKEN APPROACH

Raffin, M., Germain, E., Judd, S. (2011), Optimisation of an integrated membrane system (IMS) – A Box-Behnken approach, *Desalination*, 273, 136-141

4.1 Introduction

Increasing freshwater scarcity is making reclamation of wastewater effluent more economically attractive as a means of preserving freshwater resources. The use of an integrated membrane system (IMS), the combination of micro/ultra-filtration (MF/UF) followed by reverse osmosis (RO) membranes, represents a key process for municipal wastewater reuse; it is currently used for advanced treatment of municipal effluents for reuse in industrial processes, environmental protection/restoration, irrigation and indirect potable reuse.

A major drawback of such systems is the fouling of both the MF/UF and RO membranes. The water to be treated by the IMS system varies from one wastewater treatment plant (WWTP) to another, and its fouling propensity changes correspondingly. It is thus preferable to conduct pilot trials before implementing a full-scale plant.

Numerous studies have been performed relating to optimisation of large IMS pilot plants for membrane fouling minimisation. Studies focusing on the choice of technology include membrane type and pre-treatment (Tam *et al.*, 2007; Lozier, 2000; Park *et al.*, 2010), and operating conditions (Bartels *et al.*, 2004; Ujang *et al.*, 2007). Such studies tend to use conventional optimisation methods, varying one discrete parameter at a time, which is both time-consuming and overlooks possible synergistic effects (Khayet *et al.*, 2008). Experimental planning and/or statistical analysis are more robust process optimisation methods and widely use at laboratory scale, but less so for pilot scale plants.

Different kinds of experimental design have been applied over the years. Factorial design (Peng *et al.,* 2004; Tansel *et al.*, 2000) or fractional factorial design (Lai *et al.*, 2009; Chen *et al.*, 2003; Daramola *et al.*, 2007) are often used for optimising membrane processes. Although these experimental designs are the simplest to apply, a two-level (full or fractional) factorial design is less

comprehensive than a second-order experimental design, which then requires at least a three-level factorial design. However, an increased number of factors incurs a significant number of experiments.

The number of experiments can be reduced using alternative experimental design fitting second order models, developed over the past 50 years. These include central composite design (CDD), used by Zularisam *et al.* (2009) to optimise operating conditions for a coagulation-ultrafiltration hybrid process, and Box-Behnken design (BBD) used by Porcelli *et al.* (2009) to optimise cleaning in place of a micro-/ultra-filtration membrane. Although these experimental designs provide many advantages, especially their robustness, they are not often used at pilot scale. A *Scopus* search reveals only six membrane-based studies applying CCD (of more than 400 in the water subject area) and four studies applying BBD (of more than 150 in water) have been performed for optimising membrane processes, all at the bench-scale. These experimental designs are often associated with response surface methodology (RSM) to identify optimum operating conditions (Jokic *et al.*, 2010; Darmola *et al.*, 2007).

Other examples of optimisation based on predicting membrane fouling as a function of operating parameters and water quality include the use of an artificial neural network (ANN) (Liu *et al.*, 2009), which has previously been used for optimising forward control strategy of the MF/UF process (Cabassud *et al.*, 2002). However, the application of this model to reverse osmosis fouling prediction is still under scrutiny (Libotean *et al.*, 2009). Other models, such as hybrid system modelling, have been designed to predict the cleaning and membrane replacement interval for RO processes. However, this model is also still under development and largely theoretical (Lee *et al.*, 2009). All of these models are complex to use and none have been applied to both the MF and RO unit operations within a single treatment process.

This study demonstrates how the application of a simple experimental plan combined with statistical analysis can be used to define the operating envelope of the IMS unit operations for fouling minimisation at a large-scale pilot plant.

The study makes use of Box-Behnken Design (BBD) associated with generalised linear modelling. Whilst this method is widespread in the science field, it has not been applied to membrane process optimisation at pilot scale.

4.2 Material and methods

4.2.1 Pilot plant

To assess the technical and operational feasibility of indirect potable reuse (IPR), Thames Water has implemented a 600 $m^3.d^{-1}$ demonstration plant (Figure 4-1) in North London. The objective of this pilot plant is to provide design data for the potential implementation of a full-scale plant. The plant design parameters were identified through an extensive investigation of global IPR projects, with the aim to trial the current state-of-the-art technology (Hills *et al.*, 2007). This demonstration plant is currently treating secondary wastewater effluent and comprises a 500 µm pre-filter, a microfiltration unit, a reverse osmosis unit and an advanced oxidation process (UV + H_2O_2). Chloramine is dosed to minimise membrane biofouling, and sulphuric acid and antiscalant dosed to prevent RO membrane scaling. Permeate degassing and hydroxide dosing are used to raise the product water pH. The plant is fully automated and data recorded on a SCADA system.

The two commercial membranes studied were Memcor S10V microfiltration (MF) membranes and Hydranautics ESPA2 reverse osmosis (RO) membranes, forming part of the 600 $m^3.d^{-1}$ pilot plant. Table 4-1 summarises the membrane process specifications.

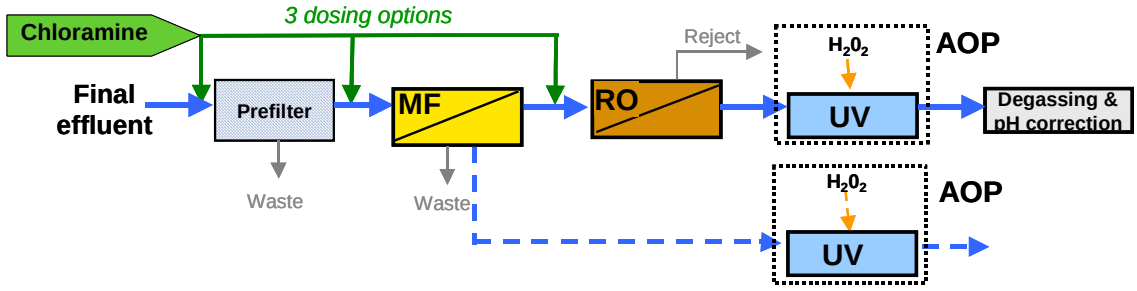


Figure 4-1 Pilot plant schematic

Table 4-1 Membrane process specifications

	Microfiltration process	*Reverse osmosis*
Manufacturer	Siemens Water Technologies Memcor Ltd	Hydranautics
Membrane Type	XS CMF-S S10V	ESPA2 (8") and ESPA2-4040 (4")
Materials	PVDF	Composite polyamide
Area/ module	25.3 m^2	ESPA2: 37.1 m^2 ESPA2-4040: 7.9 m^2
Configuration	1 cell of 16 submerged hollow fibre modules (with a possibility to increase the number of modules to 24)	3-stage with an array 1:2:1 6 spiral wound membrane modules per vessel: 1st stage: 6x8" modules 2nd stage: 12x4" modules 3rd stage: 6x4" modules
Backwash	Air + water every 15 to 45 min	None
CIP	NaOCl (540 ppm as Cl) followed by H_2SO_4 (pH 3) at 30°C	HCl (pH 2.5) at 35°C

4.2.2 Experimental plan

4.2.2.1 Assumptions

The number and range of parameters for study was limited by assuming:

(a) constant raw water quality: feed water quality was excluded as a variable.

(b) constant MF permeate water quality: the MF permeate water quality was assumed independent of both feedwater quality and operating conditions, as assumed in previous related studies (Cabassud *et al.*, 2002), allowing the MF and RO processes to be studied independently.

(c) biofouling to be negligible and/or unaffected by chloramine dosing: since no biofouling was evident on the RO membrane elements after one year of operation, chloramination dose was not studied as a variable in the RO study and it was further assumed that chloramine dosing did not influence acid and antiscalant dosing for the RO process.

4.2.2.2 Experimental plan design

For each process, four parameters were identified as potentially influencing fouling of both the MF and RO processes (Table 4-2). Combinations of parameters were determined using Box-Behnken design (Box and Behnken, 1960), decreasing the number of experiments for four parameters from 81 for a 3^n factorial design to 27 by studying the influence of parameters at three equidistant values (coded -1, 0 and +1). Table 4-3 gives the combination of levels for four parameters, each line representing four different combinations apart from the last line which is the central point of the design and is repeated 3 times.

Table 4-2 ***Parameters and their value ranges***

Param.	***MF***		***RO***	
#	***Parameters***	***Range***	***Parameters***	***Range***
x_1	Flux (LMH)	27 - 63	Flux (LMH)	15.9 - 19
x_2	BW interval (min)	15 - 45	Recovery (%)	75 - 85
x_3	Chloramine dose (ppm)	0 - 1	pH	5.9 - 7.2
x_4	Chloramine dosing point	1-3*	Antiscalant dose (ppm)	1.4 - 2.6

*1: Pre-filter, 2: Pre-MF, 3: Pre-RO

Table 4-3 Parameter combinations for a three-level Box-Behnken design, four variables

	Parameter # (from Table 1)			
Test	**x_1**	**x_2**	**x_3**	**x_4**
A	± 1	± 1	0	0
B	0	0	± 1	± 1
C	± 1	0	0	± 1
D	0	± 1	± 1	0
E	± 1	0	± 1	0
F	0	± 1	0	± 1
Validation	0	0	0	0

Each experiment (Test A to F, each with four variants and hence four experiments each) was run for seven days, or until a threshold TMP 0.7 bar was reached for the MF process or a 10% decrease in flow recorded over one stage of the RO process. For each membrane process, the volumetric ratio (Y_{exp}) was calculated of permeate produced to that projected for continuous permeate production over the course of a week. The ratio Y_{exp} was correlated with membrane fouling propensity, a ratio below unity indicating fouling.

4.2.2.3 Statistical analysis

Quasibinomial logit and quasibinomial probit generalised linear models (McCullagh and Nelder, 1989) were used to provide the model Equations (4-1) and (4-2) respectively for the MF and RO:

$$Y_{\mathrm{mod}\,el} = \frac{e^{z}}{1+e^{z}} \qquad \textbf{(4-1)}$$

$$Y_{\mathrm{mod}\,el} = \Phi(z) \qquad \textbf{(4-2)}$$

where

$$z = \beta_0 + \sum_{i=1}^{4} \beta_i x_i + \sum_{i=1}^{4} \beta_{ii} x_i^2 + \sum \sum_{i<j=2}^{4} \beta_{ij} x_i x_j) \quad \textbf{(4-3)}$$

and Y_{model} is the predicted volumetric ratio of permeate produced to the projected value for continuous permeate production, Φ the cumulative normal distribution function, and β_0 to β_i the coefficients in the polynomial function, β_{ii} the quadratic coefficients, β_{ij} the interaction coefficients, and x_i and x_j the factors to be studied - *i* taking values of 1 to 4 and *j* values of 2 to 4. The analysis was performed using the statistical software "R-Commander", and the model assessed by a chi-square test for analysis of deviance.

4.3 Results

Initial statistical analyses incorporating all parameters revealed only the backwash interval (BW) and the flux to influence MF membrane fouling rate while that of RO was influenced only by pH and recovery for the range of parameters studied (p-value<0.05 for linear, quadratic and interaction coefficient for the MF and p-value<0.05 for linear coefficient only for the RO). Chloramine dosed at 0 to 1 mg/l appeared to have no impact on MF fouling since the p-value was unity for both the chloramine dosing point and concentration. Antiscalant dosing concentration between 1.4 and 2.6 mg/l similarly produced no statistically significant change in fouling at mean fluxes between 15.9 and 19 l/(m^2.h) (or LMH) with respective a p-value of 1 and 0.33. Statistical analyses were thus repeated based on BW interval and flux as the only variables for the MF process with only linear, quadratic and interaction coefficients, and pH and recovery as the sole variables for the RO process with only the linear coefficients (Table 4-4). Equations (4-1), (4-2) and (4-3) and Table 4 provide the model Equations (4-4) and (4-5), the model equations for MF and the RO operation respectively.

Table 4-4 ***Statistical analytical results for MF and RO processe***

Parameter		*Coefficient*	*P-value*
MF			
Linear coefficients	Intercept	147	$1.14.10^{-5}$
	BW interval (min)	-2.99	$1.53.10^{-3}$
	Flux (lmh)	-2.96	$5.72.10^{-7}$
Quadratic coefficients	(BW interval (min))^2	0.01	$3.03.10^{-2}$
	(Flux (lmh))^2	0.01	$1.19.10^{-6}$
Interaction coefficients	Flux (lmh) x BW interval (min)	0.03	$3.56.10^{-4}$
RO			
Linear coefficients	Intercept	72.9	$1.32.10^{-6}$
	pH	-6.00	$1.97.10^{-7}$
	Recovery (%)	-39.4	$6.05.10^{-5}$

MF

$$Y_{Model} = \frac{\exp(147 - 2.96 \times x_1 - 2.99 \times x_2 + 0.01 \times x_1^2 + 0.01 \times x_2^2 + 0.03 \times x_1 \times x_2)}{1 + \exp(147 - 2.96 \times x_1 - 2.99 \times x_2 + 0.01 \times x_1^2 + 0.01 \times x_2^2 + 0.03 \times x_1 \times x_2)} \quad \textbf{(4-4)}$$

where x_1 is the flux (lmh) and x_2 is the backwash interval (min).

RO

$$Y_{Model} = \Phi(72.9 - 39.4 \times x_2 - 6.00 \times x_3) \quad \textbf{(4-5)}$$

where x_2 is recovery (%) and x_3 is the pH.

From Equations (4-4) and (4-5), contour plots of the predicted ratio (Y_{model}) as a function of the backwash interval and flux for the MF process (Figure 4-2), and pH and recovery for the RO process (Figure 4-3), were produced. These two figures allow the process operating envelope to be determined with reference to the range of parameters studied where membrane fouling propensity is minimised. This was applied for any parameter combination for which the Y_{model} is equal to 1. Thus, for the MF process, a flux of 32 LMH can be sustained by

backflushing every 45 minutes, whereas backflushing every 15 minutes is required to sustain operation at 56 LMH in accordance with (4-6):

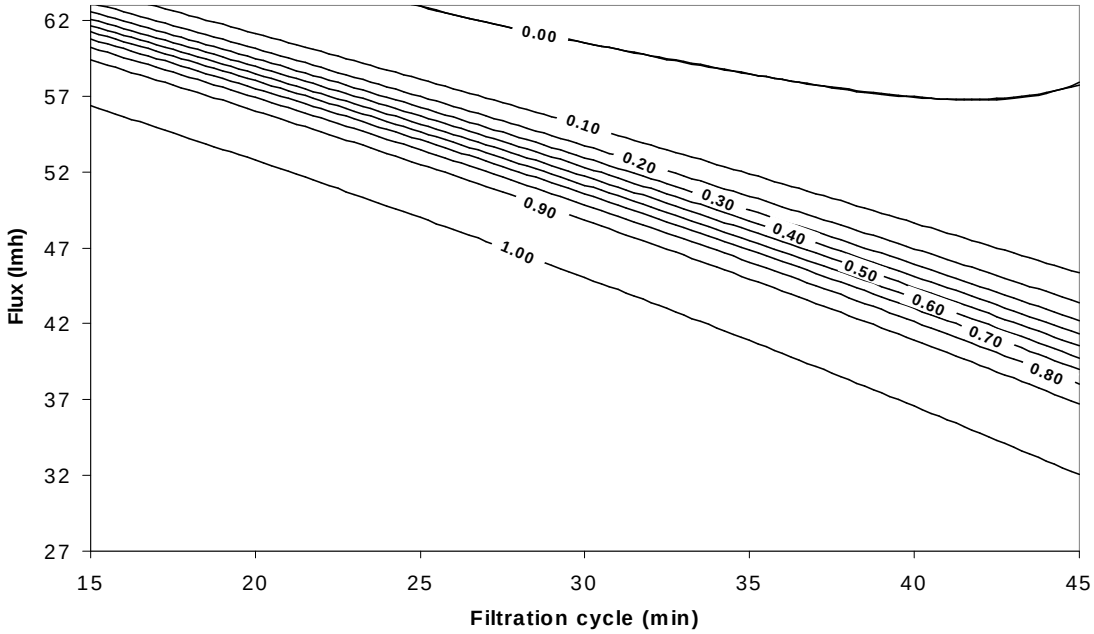


Figure 4-2 Contour plot of Y_{Model} as a function of backwash interval (min) and flux (lmh), MF

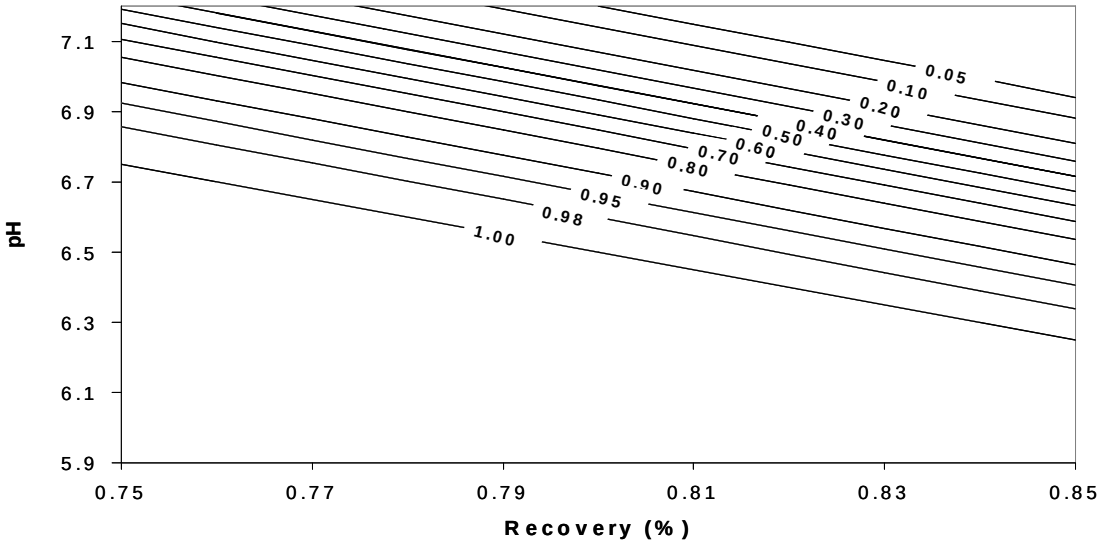


Figure 4-3 Contour plot of Y_{model} as a function of the pH and the recovery, RO

$$x_{flux} = -0.0037 \times x^{2}_{\text{BW frequency}} - 0.5859 \times x_{\text{BW frequency}} + 65.987 \quad \textbf{(4-6))}$$

For the RO, a pH of 6.25 or lower permits 85% conversion without significant fouling, whereas a recovery of 75% can be sustained for pH lower than 6.75 according to (4-7):

$$x_{pH} = -5 \times x_{\text{Recovery}} + 10.5 \quad \textbf{(4-7)}$$

This indicates that acid dosing is required since the feed water has a natural pH of ~7.2.

The model was validated by a chi-square test for analysis of deviance. This test shows that the models data fit respectively 99.9% of the experimental data for both the MF and the RO. However, the plots of Y_{Model} versus Y_{exp} (Figure 4-4 and Figure 4-5) for the MF and the RO indicate more scatter for the RO than for the MF data. This is primarily explained by superior monitoring and instrumentation on the MF plant where instrumentation (pressure meter, flow meter and water quality probes) is on-line. For the RO plant, water quality data (e.g. conductivity, pH) are recorded on SCADA. However, parameters such as flow and pressure are recorded manually several times a day. This confirms that suitable instrumentation and continuous monitoring is necessary to obtain better correlation results between the model and the experimental data.

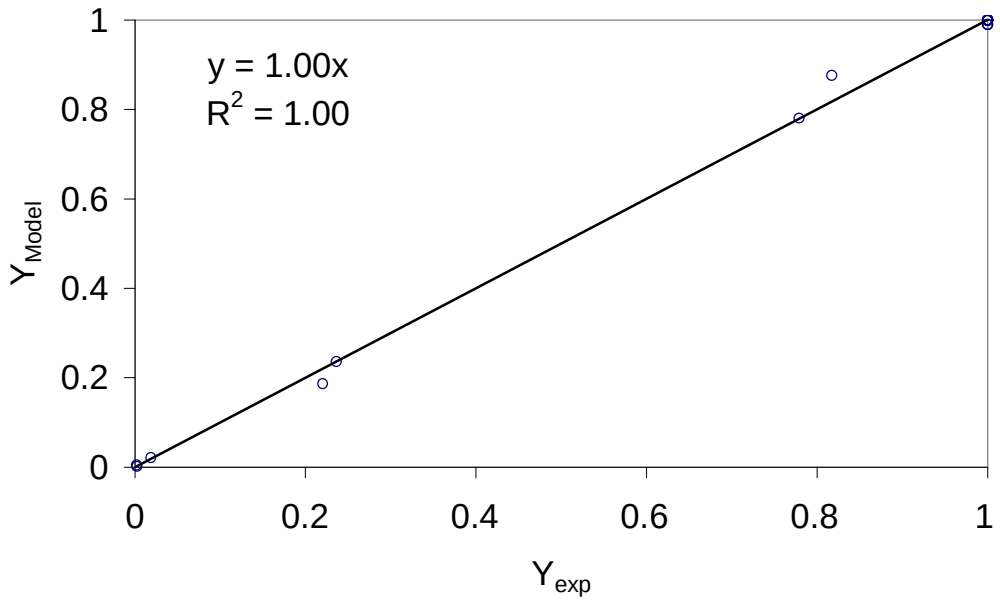


Figure 4-4 Y_{Model} **as a function of** Y_{exp} **for the MF process**

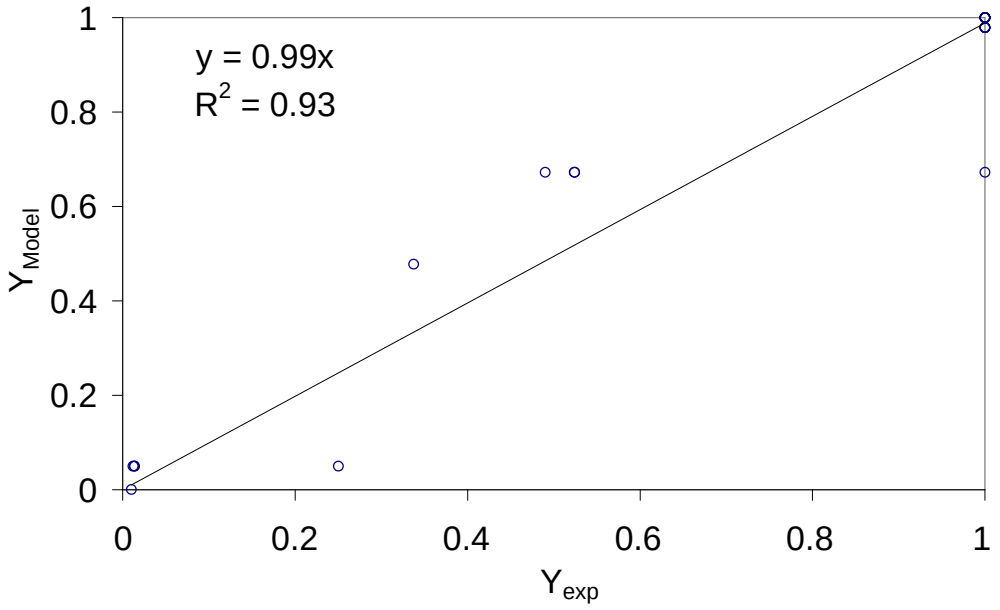


Figure 4-5 Y_{Model} **as a function of** Y_{exp} **for the RO process**

4.4 Discussion

4.4.1 Microfiltration process optimisation

Decreased fouling at lower fluxes and backwash intervals is intuitive and supported by literature data from other membrane filtration studies. Liu *et al.* (2009) showed the rate of increase in TMP for fluxes of 75 LMH (critical flux) and 150 LMH (supra-critical flux) for pressurised MF membrane to be more rapid at higher fluxes. Backwash frequencies of 30, 60 and 120 minutes were studied by Wang *et al.* (2008); these authors found that at higher backwash frequencies (30 - 60 minutes) the increase in initial TMP for each cycle, and thus irreversible fouling (i.e. fouling only removed by chemical cleaning), was lower than for lower backwash frequencies (60 - 120 min) such that higher backwash frequencies allowed longer intervals between chemical cleans. A similar conclusion was reached by Lodge *et al.* (2001), and it is generally recognised that reversible fouling is dependent on operating flux (Lin and Bérubé, 2007). At high fluxes the efficacy of regular backwashing is reduced whereas at low backwash frequencies and low fluxes increased irreversible fouling arises. This leads to consolidation of the reversible fouling onto the membrane, increasing irreversible fouling and so demanding more frequent chemical cleaning. Equation (4-6) represents the boundary between reversible and irreversible fouling.

The operating envelope determined during this study is comparable to that reported from similar processes (Table 4-5), although the ranges of fluxes and backwash frequencies applied in published studies are generally lower than the values defined by Equation (4-6). This arises because the CIP interval is greater than 21 days for the three plants. Calculation of the CIP interval based on the rate of TMP increase over the trial period for the experiment in the current study indicates that for a backwash interval of 25 minutes, the maximum applicable flux is 41 LMH based on a 21 day CIP interval.

Table 4-5 Examples of operating conditions (flux and backwash interval) for submerged MF/UF membrane processes (adapted from Wilf, 2010)

Plant	***Flux (LMH)***	***Backwash interval (min)***
Groundwater replenishment scheme, Orange county, CA	35-43	22
Kranji Newater plants, Singapore	25-33	25(20-45)
Benidorn & Rincon de Leon, Spain	35-40	22
THIS STUDY	32-56	15-45

Chloramination before the MF membrane has been widely implemented in IPR schemes worldwide. However, in this study chloramine dosing was found to have no influence on MF membrane fouling minimisation, while a decrease in chloramine concentration was observed (around 6%) between the inlet and the outlet of the MF process. Such apparent chloramine consumption by an MF process has already been reported (Thompson and Powell, 2003), but impacts of chloramine on MF membrane biofouling appear not to have been studied. Currently chloramine is dosed to inactivate bacteria, and so suppress EPS formation (extracellular polymeric substance), and oxidise the organic matter. Both actions would be expected to suppress MF membrane biofouling. However, according to Goldman *et al.* (2009), the effect of oxidising chemicals such as chloramine is limited, dependent on the nature of the organic compounds, and likely to lead to bacterial re-growth.

Water quality has not been taken into account in this study. Liu *et al.* (2009) showed water quality and operating parameters to be equally responsible for fouling of the MF membrane. Citulski *et al.* (2009) demonstrated that reversible fouling, at constant flux, can be modelled based only on total suspended solids (TSS) data and the initial TMP of each filtration cycle, irreversible fouling depending mostly on colloidal organic carbon. In the current study, it was assumed that the MF feed water quality was stable. This was shown to be reasonable by the near-constant irreversible fouling rate (measured by the increase in initial TMP for each cycle) over the course of each experiment,

indicating consistent colloidal organic carbon levels. Citulski *et al.* (2009) found intermittent increases in feed water TSS, as often manifested as turbidity "spikes", to increase reversible fouling but not the underlying irreversible fouling rate.

4.4.2 Reverse osmosis process optimisation

Four types of fouling arise on RO membranes: colloidal fouling, biofouling, organic fouling and scaling (Bartels *et al.*, 2005). Biofouling of RO membranes can be controlled by a chloramine residual in the influent (Xu *et al.*, 2010); organic fouling can be minimised by applying sufficiently low flux (Bartels *et al.*, 2005) and scaling suppressed by antiscalant dosing, pH reduction and/or reduced recovery (Ghafour, 2002). Colloidal fouling, as well as biofouling, is controlled by using MF or UF pre-treatment, and the use of coagulant before the MF/UF decreases the fouling propensity of the organic matter (Bartels *et al.*, 2005). In this study, fouling of the RO membrane, manifested as decreased permeate flow accompanied by decreased salt rejection and increased differential pressure observed in the third stage of the array, was minimised by reducing pH and recovery. Finally, the membrane permeability was completely recovered by performing an acid CIP (pH = 2.5). Based on these observations, fouling of the RO membrane was attributed primarily to inorganic scaling.

Notwithstanding this, antiscalant was found to have no influence on RO membrane fouling over the range of concentration studied. The Langelier Saturation index (LSI) indicated the saturation pH (pH_s) of the retentate water to be 6.15 for a recovery of 75% and 5.75 for a recovery of 85%. Suppression of scaling demands the solution pH to be below pH_s. Figure 4-3 implies no scaling manifested at pH levels up to 6.75 at 75% recovery and up to 6.25 at 85% recovery. This suggests that antiscalant had an influence on RO membrane scaling; antiscalant dosing associated with acid dosing has been shown to allow higher recoveries than that attained by acid dosing alone (Ghafour, 2002). However, no noticeable impact of antiscalant concentration was evident over

the narrow range of antiscalant dose studied (1.4-2.6 mg/l, as recommended by the antiscalant supplier).

Although scaling is theoretically prevented at an LSI < 0, commercial antiscalants each have threshold maximum LSI values assigned by the supplier along with the maximum solubility values for possible scalants. In this study, the maximum allowable LSI has been determined between 0.6 and 0.7 for an antiscalant dose ranging from 1.4 to 2.6 mg/l. Thus, a predicted ratio of unity implies that the LSI of the retentate is always lower than the maximum LSI allowable for the applied antiscalant. For a predicted ratio below unity the LSI of the retentate is higher than the maximum allowable LSI at some point, promoting scaling at a rate dependent on the difference between the maximum allowable and the observed LSI. The LSI change is directly proportional to the pH change and recovery; for a 1% increase in recovery, the LSI changes by 0.04.

The envelope of operating conditions determined during this study appears to be slightly more conservative (maximum recovery 85% at pH 6.25, minimum recovery 75% at pH 6.75) than the operating conditions for similar processes (Table 4-6). Higher flux and pH values have thus been applied to challenge the model. It has been found that a ratio higher than 0.98 is still required to avoid scaling; below this threshold, scaling is observed. However, a recovery of 85% at previously reported pH values could not be attained by the current plant. This arises because of the higher RO influent water quality provided by tertiary treatment at other reported sites, such as at the GWR scheme (Markus and Deshmukh, 2010) or Luggage point (Walker *et al.*, 2009) where chemical phosphorus removal is used. In the current study, the wastewater effluent underwent no tertiary nutrient removal or media filtration upstream of the plant.

Table 4-6 Examples of operating conditions (recovery and pH) for 3-stages RO membrane processes (adapted from Wilf, 2010 and Markus and Deshmukh, 2010)

Plant	***Recovery (%)***	***pH (Feed water)***
Groundwater replenishment scheme, Orange county, CA	85	7.14
Torrance,West Basin, Los Angeles County, California	85	6.4
Sulaibiya, Kuwait	85	N/A
Wollongong, Australia	85	N/A
Western Corridor projects, Australia	85	N/A
THIS STUDY	75-85	6.25-6.75

4.5 Conclusions

In this study Box-Behnken design (BBD) has been used for the optimisation of the MF and RO membranes processes. BBD was found to be a powerful statistical tool for the determination of the appropriate operating envelope for both the MF and RO, based on fouling minimisation, offering great precision (R^2=0.99 and 0.93, respectively for the MF and the RO processes, for the correlation between experimental and model data). Contrary to conventional optimisation methods, the method incorporates synergistic effects and is more robust regarding process optimisation. Moreover, although a three-level Box-Behnken design requires performing 27 experiments for 4 parameters this is still one third of the number of experiments required for a 3^n factorial design. However, a rough idea of the limits of the envelopes needs to be known before starting the experiments.

The envelope of operating conditions for the MF was found to be similar to full scale operating conditions applied for such system. As expected, lower fluxes and higher backwash frequencies reduced MF membrane fouling. Chloramine was found to have no significant effect on short term fouling of the MF process (over the course of a week). For the RO process, the envelope of operating conditions has been found to be more conservative than that usually applied to

such systems, reflecting a lower quality feedwater with respect to phosphorus concentration and temperature. As expected, lower pH and recovery values reduced RO membrane fouling. Over the narrow range of antiscalant dose studied no influence on RO membrane scaling was determined, although it was concluded that antiscalant dosing is required.

In this study it was found that there were no significant interactions between pH and the recovery for the range of parameters studied for the RO process whereas interactions between backwash interval and flux were identified for the MF process. However, the results are dependent on the range of the parameters studied; the membrane fouling cannot be predicted outside these conditions. The parameter ranges must therefore be chosen carefully since this influences the value of β_i in Equation (4-1). It is also evident that the parameter ranges determine the *p*-values for each coefficient, establishing the relationship between the parameters and the fouling propensity of the membrane. In this study, the range for the antiscalant dosing was underestimated and the results thus show no impact of this reagent.

4.6 References

Bartels C.R., Wilf M., Andes K. Iong J. (2005), Design considerations for wastewater treatment by reverse osmosis, *Water Science & Technology*, 51 (6-7), 473-482.

Bartels C.R., Franks R., Furukawa R., Murkute P., Papukchiev U. (2004), Integrated membrane system for low fouling RO desalting of municipal wastewater, final report for the Desalination Research and Innovation Partnership, conducted by the City of Oceanside in cooperation with Hydranautics.

Box G.E.P. and Behnken D.W. (1960), Some new three level designs for the study of quantitative variables, *Technometrics*, 2 (4), p. 455-475.

Cabassud M., Delgrange-Vincent N., Cabassud C., Durand-Bourlier L., Lainé J.M. (2002), Neural networks: a tool to improve UF plant productivity, *Desalination*, 145, p. 223-231.

Chen, J.P., Kim, S.L., Ting, Y.P. (2003), Optimization of membrane physical and chemical cleaning by a statistically designed approach, *Journal of membrane Science*, 219, p.27-45.

Citulski J.A., Farahbakhsh K., Kent F.C. (2009), Effects of total suspended solids loading on short-term fouling in the treatment of secondary effluent by an immersed ultrafiltration pilot system, *Water Environment Research*, 81, p. 2427-2436.

Daramola, M.O., Keesman, K.J., Spenkelink, F. (2007), Process modelling of ultrafiltration units: A RSM approach, *Journal of Applied Sciences*, 7 (23), p. 3687-3695.

Ghafour E.E.A. (2002), Enhancing RO system performance utilizing antiscalant, *Desalination*, 153, p. 149-153.

Goldman G., Starosvetsky J., Armon R. (2009), Inhibition of biofilm formation on UF membrane by use of specific bacteriophages, *Journal of Membrane Science*, 342, p. 145-152.

Hills, S., Birks, R., Grant, E., Aitken, V. (2007), Feasibility studies of planned indirect potable reuse for augmenting future water supplies in London, In: *Wastewater Reclamation and Reuse for Sustainability 2007 Conference*, Antwerp, Belgium, 9-11 March 2007.

Jokic, A., Zavargo, Z., Seres, Z., Tekic, M. (2010), The effect of turbulence promoter on cross-flow microfiltration of yeast suspensions: A response surface methodology approach, *Journal of Membrane Science*, 350, p. 269-278

Khayet M., Cojocaru C., Zakrzewska-Trznadel G. (2008), Response surface modelling and optimisation in pervaporation, *Journal of Membrane Science*, 321, p. 272-283.

Lai, W-L., Chen, L-F., Chen, J-J., Liao, S-W. (2009), Effects of operational parameters on carobon recovery and water flux in ultrafiltration using fractional factorial design, *Desalination*, 249, p. 1365-1370.

Lee Y-G., Gambierb A., Badreddinb E., Leec S., Yangd D.R., Kim J.H. (2009), Application of hybrid systems techniques for cleaning and replacement of a RO membrane, *Desalination*, 247, p. 25-32.

Libotean, D., Giralt, J., Giralt, F., Rallo, R., Wolfe, T., Cohen, Y. (2009), Neural Network approach for modelling the performance of reverse osmosis membrane desalting, *Journal of Membrane Science*, 326, p. 408-419.

Lin H. and Bérubé P.R. (2007), Modelling the impact of permeate flux and hydrodynamic conditions on fouling in submerged hollow fiber membranes, *Water Science & Technology: Water Supply*, 7(4), p. 111-118.

Liu Q-F., Kim S-H., Lee S. (2009), Prediction of microfiltration membrane fouling using artificial neural network models, *Separation and Purification Technology*, 70, p. 96-102.

Lodge, B.N., Judd, S.J., and Smith, A.J. (2001), A statistical method for quantifying the different fouling effects of three combined water sources on an ultrafiltration membrane, *Desalination*, 142, p. 143-149 (2001).

Lozier, J. (2000), Two approaches to indirect potable reuse using membrane technology, *Water Science and Technology,* 41 (10-11), p. 149-156.

Markus, M.R., Deshmukh, S.S. (2010), An innovative approach to water supply – the groundwater replenishment system, In: *World Environmental and Water Ressources Congress 2010: Challenges of Change*, Providence, Rhode Island 16-20 May 2010, p. 3624-3639.

McCullagh P. and Nelder J.A. (1989), *Generalized linear model*, 2nd Ed., Chapman & Hall, p. 13-14.

Park, C., Hong, S-W., Chung, T.H., Choi, Y-S. (2010), Performance evaluation of pre-treatment processes in integrated membrane system for wastewater reuse, *Desalination*, 250, p. 673-676.

Peng, W.. Escobar, I.C., White, D.B. (2004), Effect of water chemistries and properties of membrane on the performance and fouling - a model development study, *Journal of Membrane Science*, 238 (1-2), 33-46.

Porcelli, N., Hillis, P. and Judd, S. (2009a), Microfiltration membrane plant start up: A case study with autopsy and permeability recovery analysis, *Environmental Technology*, 30 (6), p. 629-639.

Tam, L.S., Tang, T.W., Lau, G.N., Sharma, K.R., Chen, G.H. (2007), A pilot study for wastewater reclamation and reuse with MBR/RO and MF/RO systems, *Desalination*, 202, p. 106-113.

Tansel, B., Regula, J., Shalewitz, R. (2000), Evaluation of ultrafiltration process performance for treatment of petroleum contaminated waters, *water, Air, and Soil Pollution*, 126, p. 291-305.

Tercero Espinoza L.A., Rembor M., Arriba Matesanz C., Heidt A., Frimmel F.H. (2010), Formation of bromoform in irradiated titanium dioxide suspensions with varying photocatalyst, dissolved organic carbon and bromide concentration, *Water Research*, 43, p. 4143-4148.

Thompson M. and Powell D. (2003), Case study – Kranji high grade water reclamation plant, Singapore, *IMSTEC'03*, September 2003, Australia

Ujang, Z., Ng. K.S., Hamzah T.H.T., Roger P, Ismail M.R., Shahabudin S.M, Abdul Hamid M.H. (2007), Application of immersed MF (IMF) followed by reverse osmosis (RO) membrane for wastewater reclamation: A case study in Malaysia, *Water Science and Technology*, 56 (9), p. 103-108.

Wang L., Wang X., Fukushi K-I. (2008), Effects of operational conditions on ultrafiltration membrane fouling. *Desalination*, 229, p. 181-191.

Wilf, M. (2010), The guidebook to membrane technology for wastewater reclamation, 1st Ed., Balaban Publisher Publications, Hopkinton, USA.

Xu P., Bellona C., Drewes J.E. (2010), Fouling of nanofiltration and reverse osmosis membranes during municipal wastewater reclamation: Membrane autopsy results from pilot-scale investigations, *Journal of Membrane Science*, 353, p. 111-121.

Zularisam, A.W., Ismail, A.F., Salim, M.R., Sakinah, M., Matsuura, T. (2009), Application of coagulation-ultrafiltration hybrid process for drinking water treatment: Optimization of operating conditions using experimental design, *Separation and Purification Technology*, 65 (2), p. 193-210.

CHAPTER 5: BACKWASH AND FLUX OPTIMISATION FOR WASTEWATER REUSE USING MICROFILTRATION

Raffin, M., Germain, M., Judd, S. (2011), Backwash and flux optimisation for wastewater reuse using microfiltration, Submitted to *Water Research*

5.1 Introduction

Microfiltration (MF) and ultrafiltration (UF) membranes are widely used for pre-treatment of reverse osmosis (RO) processes in wastewater recovery for Indirect Potable Reuse (IPR) (Lazarova *et al.*, 2008). However, a major drawback of such systems is the fouling of the MF/UF membrane which considerably reduces the process throughput.

Membrane fouling is determined both by feedwater quality and process operating parameters; numerous studies have been performed regarding the identification of membrane foulants in wastewater effluent treatment (Zheng *et al.*, 2009; Citulski *et al.*, 2009; Jarsutthirak *et al.*, 2002). Fouling is usually defined as reversible, if removed by physical cleaning such as backwashing, or irreversible if removed only through the application of chemicals. Whist precise foulant speciation is abstruse it is often generally be categorised as “particulate” or “organic”, these are obviously not mutually exclusive.

Particle concentration, as total suspended solids (TSS), has been shown to proportionally diminish membrane flux and increase transmembrane pressure (TMP), over the filtration cycle (Citulski *et al.*, 2009; Bourgeous *et al.*, 2001). Bourgeous *et al.* (2001) also showed irreversible fouling to become prevalent at higher TSS concentrations, where higher TSS loads on the membrane were not completely removed by physical cleaning. Moreover, smaller particles have been found to more tenaciously adhere to the membrane surface, since the shear forces they are subject to are lower than those of larger particles; on a mass basis fouling resistance has been reported to increase by 50% with a five-fold decrease in particle diameter (Bourgeous *et al.*, 2001). Whilst fouling by particles is normally associated with cake formation at particle sizes below that of the membrane pore size, pore blocking may also take place (Mousa and Al-Hitmi, 2007; Hwang *et al.*, 2009); Hwang *et al.* (2009) demonstrated that internal membrane fouling can contribute significantly more to total filtration resistance than the cake layer. Thus, whilst particle fouling is generally

expected to be removed by physical cleaning (Psoch and Shiewer, 2006), the more tenacious and generally smaller particles apparently contribute most significantly to irreversible fouling.

Organic fouling is generally understood to comprise colloidal and dissolved organic material, and thus may be differentiated from particulate fouling only by size for entirely organic feedwater constituents. Several studies have sought to identify the constituent primarily responsible for membrane fouling through employing fractionation. Jarsuutthirak *et al.* (2002) concluded that organic hydrophilic colloids of >3500 Dalton size range, such as polysaccharides, contributed more to fouling of PA-UF membranes than the hydrophobic (humic and fulvic acid) and transphilic fractions. Zheng *et al.* (2009) found dissolved organic compounds in the 0.45 – 0.26 μm size range to provide the highest organic fouling propensity compared to large colloids (>0.45 μm) and components smaller than the UF pore size; these dissolved organic compounds were identified as biopolymer.

Fouling by organic matter is mechanistically more complex than that by particles. The fouling mechanism associated with natural organic matter (NOM), for example, depends upon NOM heterogeneity, membrane type, pH, ionic strength and multivalent cations concentration (Cho *et al.*, 2000; Lee *et al.*, 2004). Extracellular polymeric substances (EPS), generated through microbiological activity, enhance the attachment of bacteria to the membrane and so significantly contribute to irreversible fouling; EPS and soluble microbiological compounds from secondary treatment have been observed as being the major contributor to the gel layer (Nguyen et al, 2009).

Membrane fouling can be suppressed by operation under sub-critical conditions, i.e. under conditions sufficiently benign for fouling to be significant (Bacchin and Aimar, 2005). Critical fouling conditions may be defined with respect to the operating flux (Bacchin *et al.*, 2006; Le-Clech *et al.*, 2006) and/or the amount of material. The latter include the filtered volume between physical cleans (McAdam and Judd, 2008), the solute osmotic pressure (Bessiere *et al.*,

2005) or the critical deposit formation related to the contaminant mass transfer (Bacchin, 2004). This critical condition can be identified experimentally by plotting the TMP, fouling rate or other fouling index as a function of the condition, criticality being observed as a deviation from linearity.

Whilst progress continues to be made in elucidating fouling mechanisms pertaining to the foulant character, studies of operating conditions impacts have been largely constrained to flux and have been conducted mainly at laboratory scale. It has been acknowledged that such studies do not fully capture the water quality conditions typically encountered during wastewater treatment plant operation, or full-scale membrane module properties such as fibre length and packing density (Citulski *et al.*, 2009). The aim of this study is to determine fouling behaviour under conditions replicating those of a full-scale plant. Specifically, effects of both operating parameters (flux and backwash interval) and water quality (turbidity and temperature) on irreversible and reversible fouling rates are reported as pertaining to critical fouling conditions and chemical cleaning intervals.

5.2 Materials and Methods

5.2.1 Microfiltration unit and pilot plant overview

The pilot plant has been described elsewhere (Raffin *et al.*, 2011). The MF technology (Memcor *CMF-S*, Table 5-1) was supplied by Siemens. The skid employed 24 *S10V* hollow fibre modules, forming part of a 600 $m^3.d^{-1}$ demonstration plant treating secondary wastewater effluent and including a 500 µm pre-filter, the microfiltration unit, a reverse osmosis (RO) unit and an advanced oxidation process (AOP). The plant was fully automated and data recorded on a SCADA system. The average water quality (measured online) of the MF feedwater for 2008-2010 is reported in Table 5-2.

Table 5-1 Membrane module specifications

Manufacturer	Siemens Water Technologies Memcor Ltd
Membrane Type	XS CMF-S S10V
Materials	PVDF, 0.04 µm pore size
Area/module	25.3 m^2
Configuration	1 cell of 24 submerged hollow fibre modules
Filtration mode	Out-in
Design flux	27-37 LMH
Design Backwash	Backwash interval: 15-45 min Air (0.40 m/h for 55 s) + water (0.06 m/h for 15 s). Backwash downtime 300 s
Operating temperature	>0-40ºC (max 45ºC)
Operating pH	2-10.5
Standard CIP	Recommended interval of 15 days 600L NaOCl solution (540 ppm, 30ºC) followed by 600L H_2SO_4 solution (pH 2.5, 30ºC)

Table 5-2: Average feed water quality (2008-2010)

Parameter	Average	Min	Max
Turbidity (NTU)	6.18 ±3.35	0.37	100
TOC (mg/L)	7.18±0.82	5.82	8.88
Temperature (^oC)	16.7 ± 1.97	8.56	26.54
pH	7.09 ± 0.35	6.55	7.85
Conductivity ($\mu S.cm^{-1}$)	1048 ± 90	630	1862
UV_{254}	0.196±0.018	0.175	0.256
Specific UV absorbance ($m^{-1}.mg^{-1}.L$)	2.82±0.45	2.14	4.35

5.2.2 Data acquisition, collation and analysis

18 months of data from the MF unit operation were processed, relating to membrane fouling as influenced by plant operating parameters and feedwater quality. Data included flow and TMP, from which operating fluxes and permeabilities were calculated. Fluxes were normalised to a temperature of 20^oC according standard methods (EPA 815-R-06-009). Turbidity, pH, conductivity and temperature were monitored on-line, and UV_{254} measured off-line using a Genesys *10uv* spectrophotometer. The MF reversible and irreversible fouling rates (i.e. rate of change of TMP under constant flux conditions) were calculated for each filtration cycle, i.e. the cycle of 15-45 minutes between backwashing (BW), and each cleaning cycle, the cycle of 15-21 days between each cleaning in place (CIP), over the entire 18 month period.

The TMP values recorded at the start of each filtration cycle were plotted against time, over the course of a single cleaning cycle, and the irreversible fouling rate determined from the slope. This was applied to each of the 32 cleaning cycles over the course of the 18 month study. A macro was used to calculate the reversible fouling rate, this being the rate of TMP increase during a single filtration cycle, for all 32760 filtration cycles. Corresponding feedwater quality data, and specifically temperature and turbidity, were also recorded. The subsequent fouling rate data collated for the entire operational period were then sorted by flux, turbidity and temperature. Mean reversible fouling rates were determined for those data having common values of feedwater turbidity (± 1 NTU) and temperature (± 2.5 oC), and the standard deviation determined.

5.3 Results and discussion

Results show the mean irreversible fouling (i.e. fouling only removed by chemical cleaning) to depend both on flux and backwash interval (Figure 5-1), whereas reversible fouling apparently depended only on flux (Figure 5-2). Irreversible fouling at 63 LMH and a 45 min BW cycle could not be measured during the experiments since the fouling rate was sufficiently high for the process to be automatically shutdown on exceeding the threshold maximum pressure of 0.7 bar.

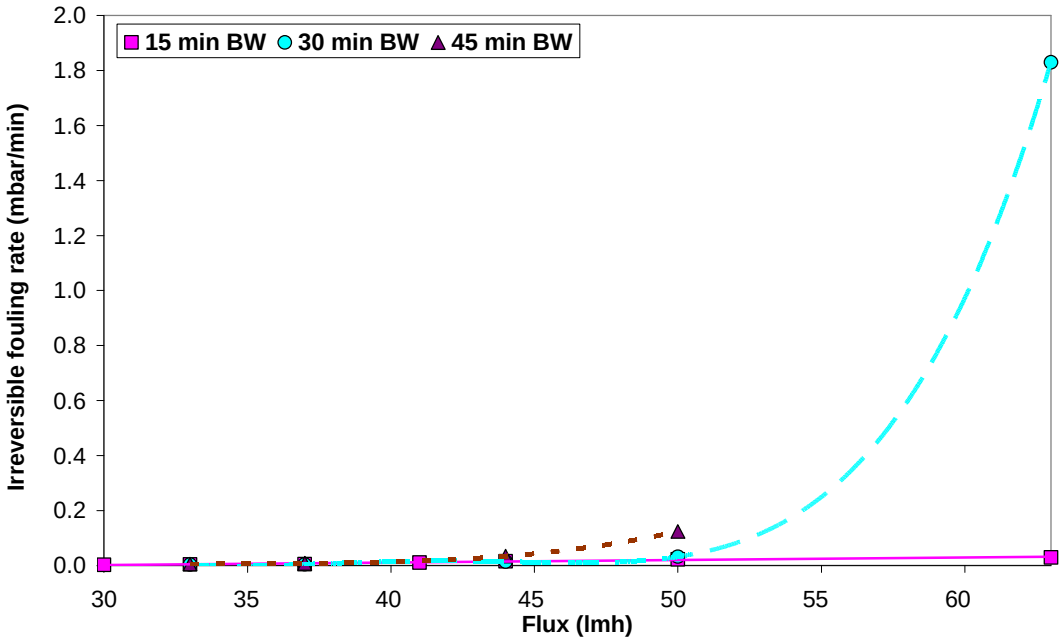


Figure 5-1: Irreversible fouling rate as the function of the flux at backwash intervals of 15, 30 and 45 min

Decreased fouling at lower fluxes and backwash intervals is intuitive and supported by literature data from previous studies. Liu *et al.* (2009) showed the rate of TMP increase for fluxes of 75 LMH (the critical flux) and 150 LMH (supra-critical flux) for a pressurised MF membrane to be more rapid at the higher flux by 10 to 70% depending on water quality. Backwash intervals of 30, 60 and 120 min were studied by Wang *et al.* (2008), who found that at higher backwash interval (30–60 min) the increase in initial TMP for each filtration cycle, and thus irreversible fouling, was lower than that at lower backwash interval (60–120 min). In the study of Wang *et al.* (2008), the time to reach the threshold TMP was found to decrease by 25% when the backwash interval was increased from 60 to 120 min.

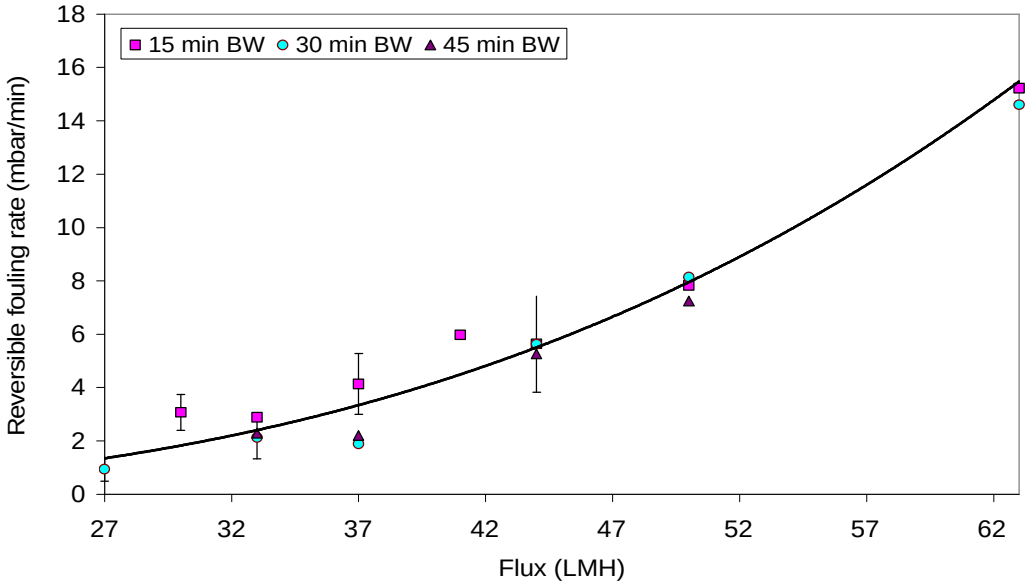


Figure 5-2: Reversible fouling rate as the function of the flux at backwash intervals of 15, 30 and 45 min

Shorter backwash intervals thus allow longer intervals between chemical cleans, due to suppression of reversible fouling, whilst the contribution from irreversible fouling has been reported to increase with increasing operating flux (2007). At high fluxes and/or longer backwash intervals, backwash efficacy is apparently reduced, since reversible fouling becomes consolidated and more irreversible and so demanding more frequent chemical cleaning.

5.3.1 Water quality

Water quality is obviously a key parameter determining fouling propensity of the membrane, with turbidity and UV_{254} shown to be as important as flux and backwash interval in determining MF fouling (Liu *et al.*, 2009). In the current study the TOC concentration and UV_{254}, associated with soluble and colloidal organic matter, did not vary by more than 10% (Table 5-2). This is reflected in the near-constant irreversible fouling rate, represented by the increase in initial

TMP for each cycle, over the course of the trial. However, it was also observed that rapid increases in turbidity could lead to a shutdown of the MF process due to the threshold TMP being exceeded. Irreversible fouling did not correlate with turbidity, since the initial TMP for each filtration cycle was independent of turbidity fluctuations (Figure 5-3). Conversely, the development of reversible fouling followed the same sinusoidal pattern as turbidity, corroborating reported information from Citulski *et al.* (2009). These authors found intermittent increases in feed TSS, manifested as turbidity "spikes", to increase reversible fouling without affecting the underlying irreversible fouling rate.

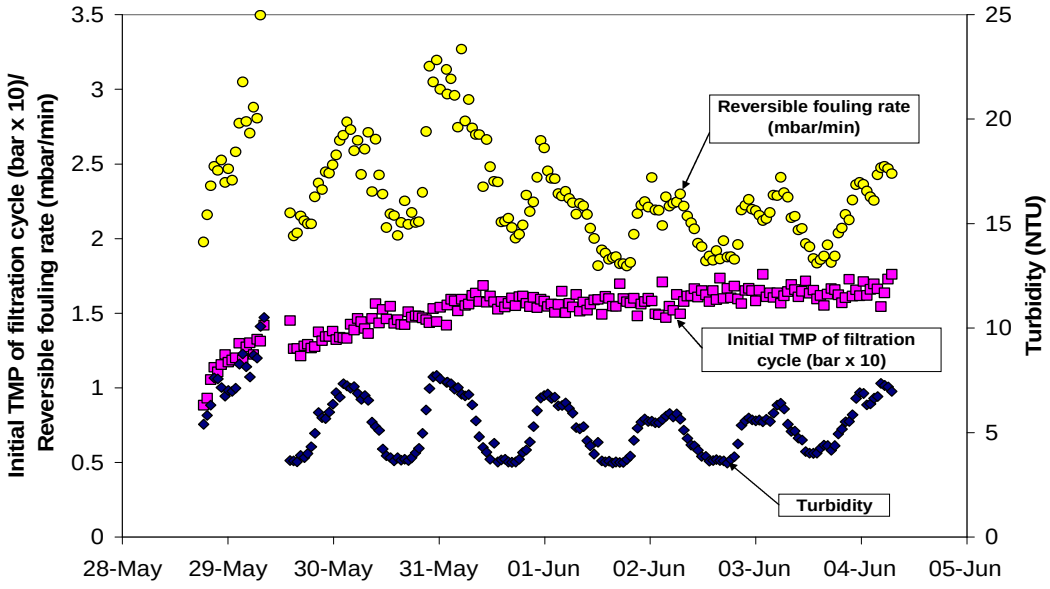


Figure 5-3: Initial TMP of filtration cycle, reversible fouling rate and turbidity as a function of the time

Reversible fouling rates plotted as a function of flux at constant turbidity and temperature (Figure 5-4) revealed a neo-exponential relationship (R^2 = 0.89-0.99), as assumed in most of previous studies of fouling rate trends with flux (Ognier *et al.*, 2002; Le-Clech *et al.*, 2003; Brookes *et al.*, 2006). Many of such reported trends relate to membrane bioreactors (MBRs), however. Whilst the correlation coefficient was reasonably high for the exponential trend in the

current study, a consistent power relationship was found to be slightly more consistent (R^2 = 0.93-0.99). Mean reversible fouling rates plotted as a function of turbidity at fluxes of 27 to 63 LMH and constant temperature (Figure 5-5) showed fouling to increase exponentially with turbidity, yielding correlation coefficients of 0.92-0.99 (Table 5-3). Such trends are in accordance with that reported in MBR studies by Li *et al.* (2010), who studied the influence of TOC concentration on reversible fouling rate, and Brookes *et al.* (2006), who reported impacts of mixed liquor suspended solids concentration on fouling rate.

The impact of solids concentration, as represented by turbidity in the current study, on reversible fouling rate has been reported in a number of studies, with reference to concentrations of particles (Citulski *et al.*, 2009; Bourgeous *et al.*, 2001), biopolymers or general natural organic matter (Zheng *et al.*, 2009), and other materials such as whey or BSA proteins (Carić *et al.*, 200; She *et al.*, 2009). From Figure 5-5 and Table 5-3, it is evident that reversible fouling rate is non-zero and flux-dependant at a zero turbidity. This zero-turbidity reversible fouling rate, as extrapolated from Figure 5-5 data, is included in Figure 5-4 and indicates the same trend in flux as the finite turbidity data. Thus, whilst reversible fouling apparently follows turbidity (Figure 5-4), it is apparent that other constituents contribute to this fouling. Zheng *et al.* (2009) reported the 0.026-0.45 µm size fraction to contribute more significantly to resistance than the >0.45 µm suspended solids size fraction. The contribution of non-turbid matter to overall cake resistance can be calculated from Figure 5-4. The resulting trend (Figure 5-6) indicates a linearly increasing contribution from this material at decreasing turbidity – from 50% at 10 NTU to 85% at zero turbidity. The resistance offered by this foulant fraction thus remains unchanged, whilst cake resistance nonetheless increases with turbidity overall. It is thus apparent that the non-turbid water constituents contributed both to reversible and irreversible fouling in this instance.

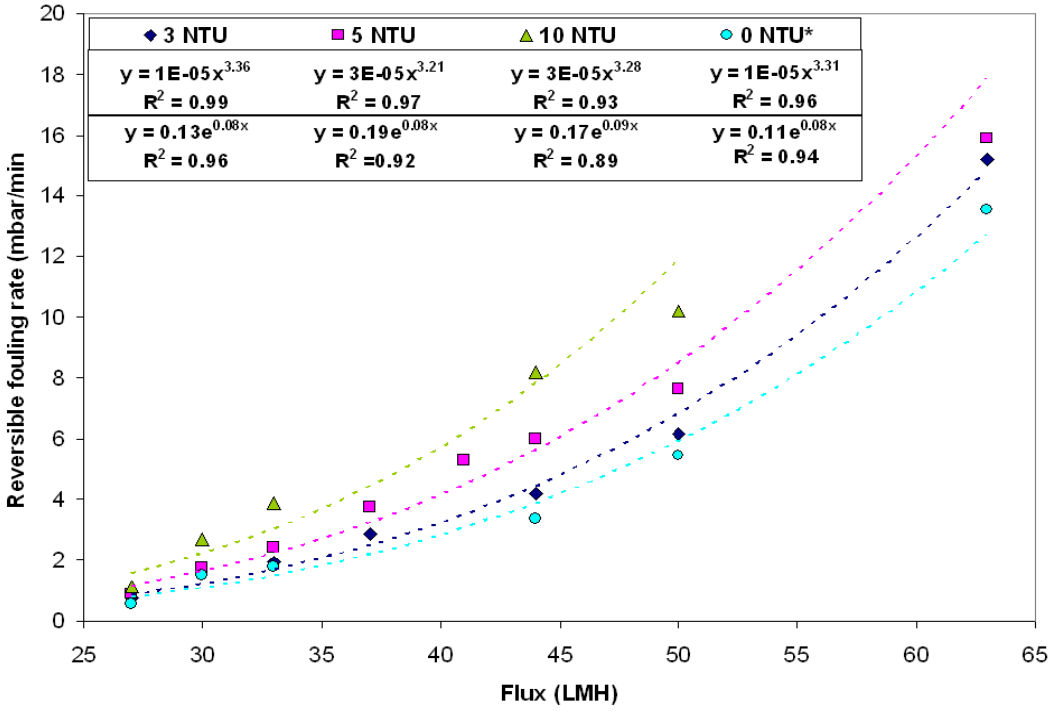


Figure 5-4: Reversible fouling rate as the function of flux at constant turbidity and constant temperature (data extrapolated from Figure 5-6)

Table 5-3: Coefficients *a* and *b* and correlation factors of *Reversible fouling rate= a e^(b flux)*

Flux (LMH)	*a*	*b*	R^2
27	0.56	0.086	0.98
30	1.48	0.046	0.95
33	1.78	0.064	0.98
37	1.69	0.149	0.99
44	3.37	0.074	0.92
50	5.44	0.061	0.96
63	13.53	0.031	0.98

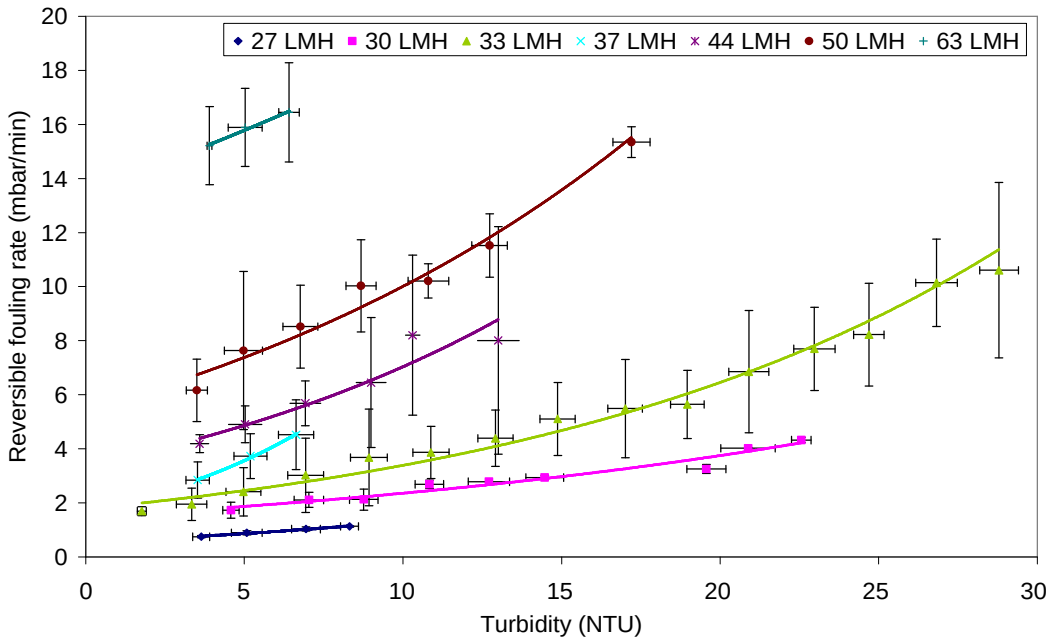


Figure 5-5: Reversible fouling rate as a function of the turbidity for different fluxes and constant temperature (15±2.5 °C)

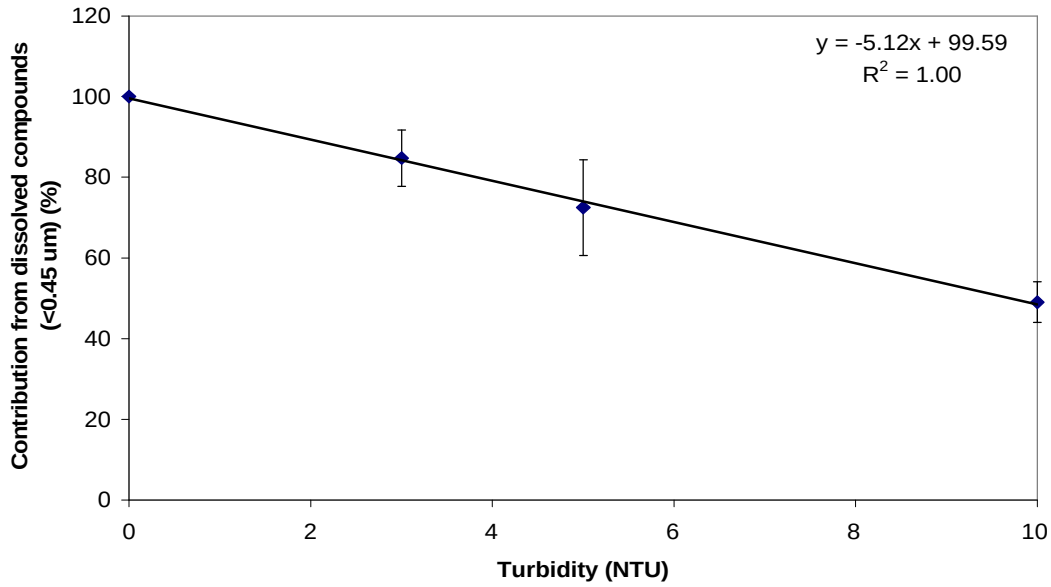


Figure 5-6: Contribution to cake resistance from non-turbid matter (%) as a function of the turbidity (NTU)

5.3.2 Temperature

The reversible fouling rate was found to be higher at lower temperature despite normalising the viscosity to 20^{o}C (Figure 5-7), an exponential relationship again describing change in fouling rate with temperature. This once again supports previous findings, and has been variously attributed to increased membrane pore rigidity at lower temperatures for PES membranes (Amin *et al.*, 2010) and the membrane polymeric structure (Wang, 1988). Wang (1988) reported a non-linear relationship between flux and inverse viscosity, conflicting with classical Newtonian viscosity-based resistance relationships. It was concluded that for low viscosity liquids an infinitely small increase in viscosity could bring about an abrupt decrease in permeability, attributed by this author to the differing membrane structure to that of natural porous materials. A decrease in temperature also reduces foulant back transport away from the membrane under Brownian diffusive forces (Jiang *et al.*, 2005).

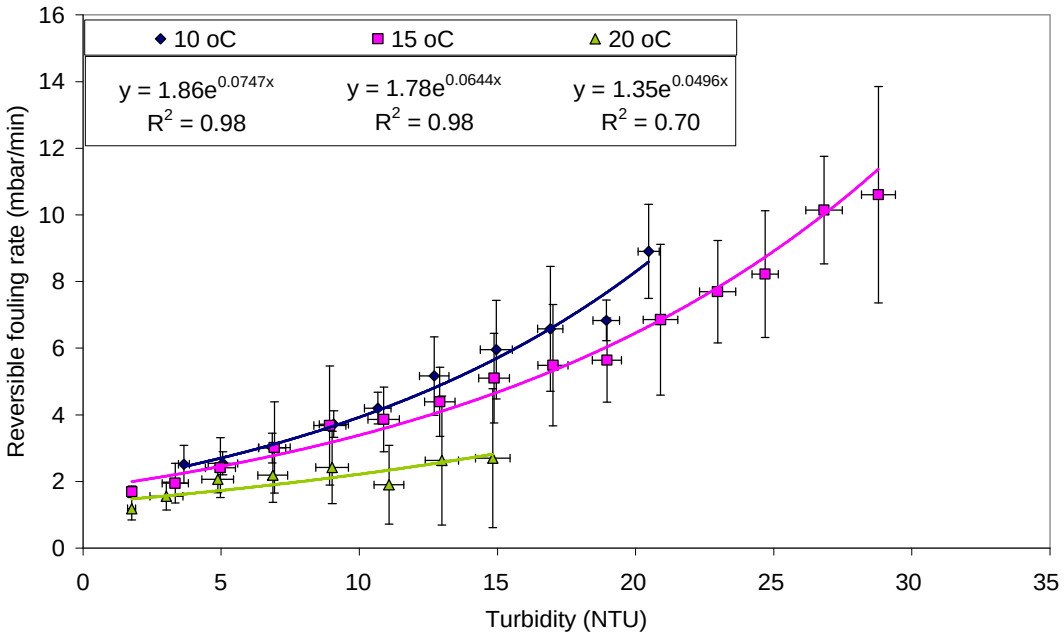


Figure 5-7: Reversible fouling rate as a function of turbidity at different temperatures (Flux: 33 LMH)

5.3.3 Irreversible fouling: CIP interval

The interval between chemical cleans ($CIP_{interval}$) is given by:

$$CIP_{interval} = \frac{TMP_{max} - TMP_0 - K_{reversible} BW_{frequency}}{K_{irreversible}} \quad \textbf{(5-1)}$$

where TMP_{max} is the maximum transmembrane pressure of operation, 500 mbar in this case, TMP_0 is the initial TMP after the CIP, $K_{reversible}$ and $K_{irreversible}$ are the reversible and irreversible fouling rates mbar.min^{-1}, and $BW_{interval}$ is the backwash interval in minutes. The maximum TMP is often that recommended by the manufacturer either to minimise irreversible deposition of foulants or/and cavitation of the filtrate pump. However, this value is often conservative.

The CIP interval can be represented as a function of flux at different backwash intervals but constant turbidity and temperature (Figure 5-8), or as a function of turbidity at different temperatures but constant flux and backwash interval (Figure 5-9). Figure 5-8 shows that at low flux and shorter backwash intervals the latter contributes little to the CIP interval. However, as flux increases the backwash interval becomes increasingly significant: at the highest backwash interval explored, a significant decrease in CIP interval (e.g. 26% at 33LMH) arises, even at low flux. A flux increase also yields a decreased CIP interval, e.g. 73 to 86% on increasing the flux from 37 LMH to 44 LMH for a backwash interval of 15 to 45 min, as indicated in previously reported work (Wang *et al.*, 2008; Lin and Berubé, 2007). Figure 5-9 indicates that increased turbidity and decreased temperature decreases the CIP interval, but that CIP interval is unaffected by temperatures between 10 and 20°C at turbidities <7 NTU.

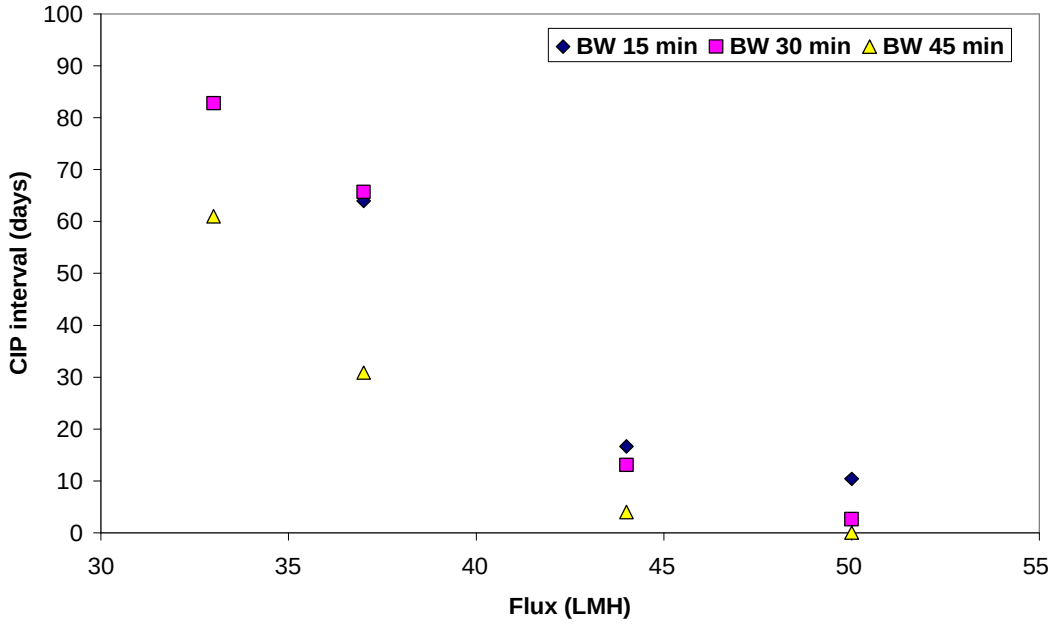


Figure 5-8: CIP intervals (days) as a function of the flux (LMH) at different backwash interval (Turbidity: 5±1 NTU, Temperature 15±2.5 ^{o}C)

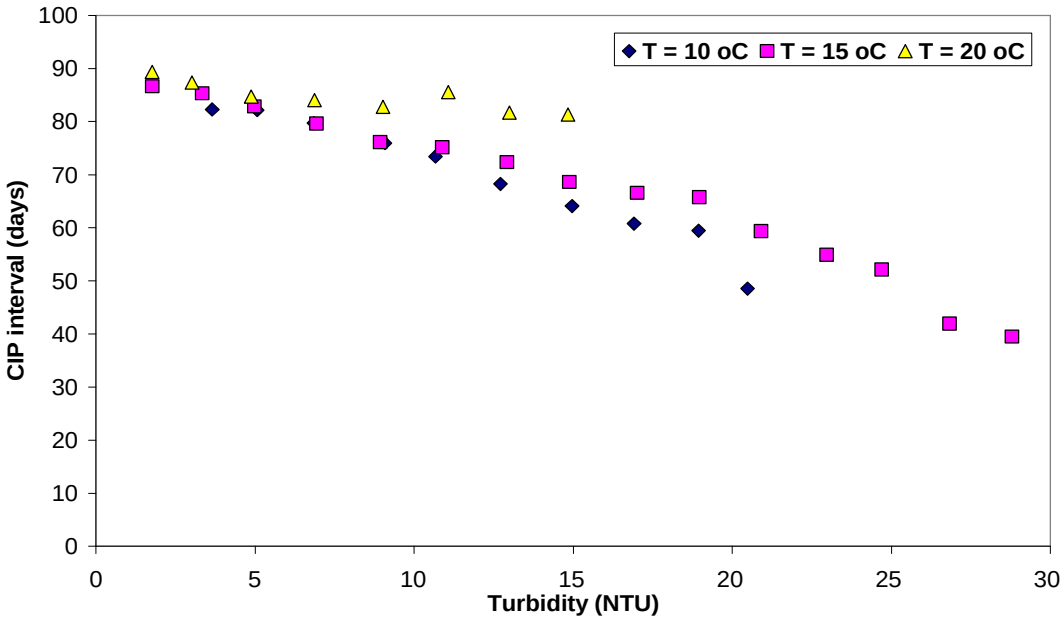


Figure 5-9: CIP intervals (days) as a function of the turbidity at different temperatures (Flux: 33 LMH, BW interval: 30 min)

Figure 5-10 shows the evolution of the CIP interval with flux for different maximum TMPs. The figure indicates that CIP interval can be increased by 74% at 33 LMH and TMP_{max} = 700 mbar, and by 500% at 44 LMH and 860 mbar compared with operation at a TMP_{max} of 500 mbar. However, an increase in TMP_{max} implies an increased permeate pumping energy demand. Monitoring of the energy consumption of the permeate pump at different fluxes and TMP revealed that, on average, the OPEX increased by up to 8% on increasing TMP_{max} to 700 mbar and by up to 20% on increasing it to 860 mbar for constant flux and constant backwash frequency, similar to previous reports (Parameshwaran *et al.*, 2001).

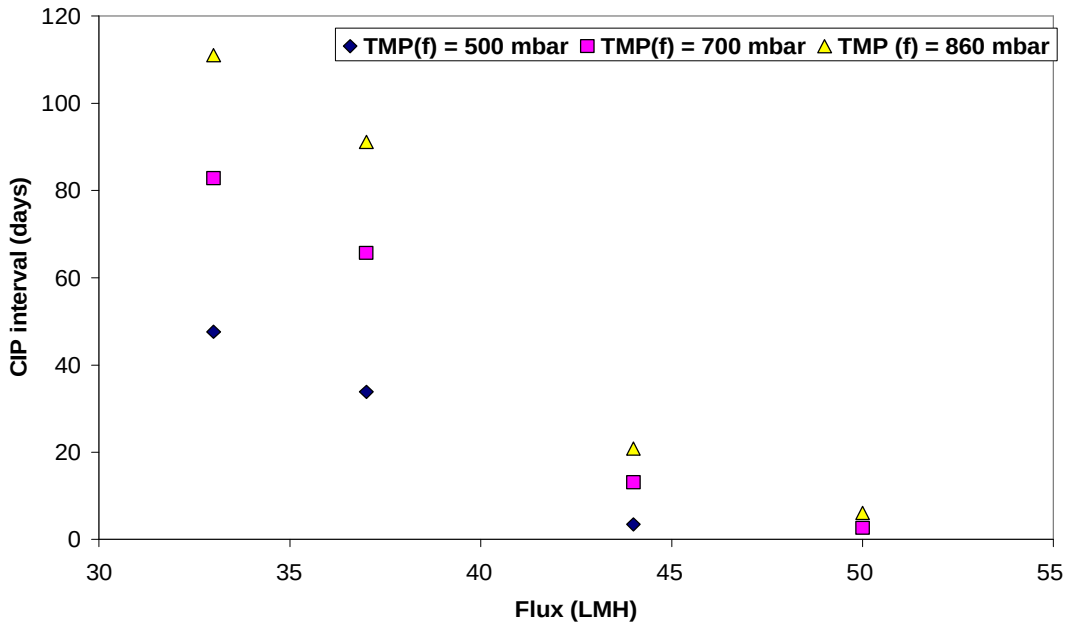


Figure 5-10: CIP interval (days) as a function of flux (LMH) for different maximum TMP

5.3.4 Modelling

An empirical model to define operating conditions (flux and backwash interval) as a function of water quality (turbidity and temperature) for a fixed clean-in-

place interval ($CIP_{interval}$) was attempted. However, no model equation could be defined, either by entering the $CIP_{interval}$ or the irreversible and reversible fouling rates as dependent variables (i.e. by defining equations for $K_{reversible}$ and $K_{irreversible}$ in Equation (5-1)). This arises from the imbalanced distribution of data leading to an excessive determined significance of both flux and temperature on reversible fouling rates, such that backwash interval and turbidity become insignificant factors which is in contravention with the experimental data.

According to the definition of critical flux given by Bacchin *et al.* (2006), data from the current study indicate that at short backwash intervals the critical flux with respect to irreversible fouling is not reached at the highest flux of 63 LMH studied (Figure 5-2). However, increasing the backwash interval depresses the critical flux, to 49 LMH at a 30 minute backwash interval and 39 LMH at an interval of 45 minutes. For reversible fouling, it appears that even at turbidities below 3 NTU (Figure 5-5) the critical flux for development of significant reversible fouling is less than 27 LMH. However, it is clear that for each flux a critical turbidity, which can be associated with the suspended solids concentration, can be defined (Figure 5-5) which suppresses the flux. For example the critical turbidity values at 30 and 33 LMH are respectively 10 and 5 NTU. Figure 5-8 also shows that the critical concentration depends on temperature. It appears, therefore, that no unique critical flux can be defined for real wastewater since it depends on both the water quality and backwash efficiency.

The concept of sustainable flux (Bacchin *et al.*, 2006), defined as the flux at which the process is sustainable in terms of operation and costs (OPEX and CAPEX), is more appropriate when optimising a real process. According to Figure 5-8 to Figure 5-10, even at fluxes above the so-called critical flux and at any turbidity, a longer CIP interval can be attained. For example, for a 30 day CIP interval, fluxes up to 41 LMH and 37 LMH can be respectively applied for a 15-30 min backwash interval and 45 min backwash interval. These two fluxes are, however, at the upper limit or higher than the fluxes recommended by the supplier.

5.4 Conclusions

Reversible and irreversible fouling rates for a microfiltration membrane treating secondary wastewater effluent have been measured over each filtration cycle and between each chemical clean in place (CIP). Fouling rates were correlated with membrane plant operating parameters and water quality parameters after applying the standard Newtonian viscosity correction.

Irreversible fouling was found to be dependent on both operating flux and backwash interval, but did not vary with measured water quality parameters. Reversible fouling, however, was independent of the backwash interval and dependent on operating flux, turbidity and temperature. For both irreversible and reversible fouling, an increase in flux, backwash interval and turbidity and a decrease of temperature led to an increase in both fouling rates, with temperature-correction viscosity proving insufficient to account for the influence of temperature. Power or exponential functions were found to describe the relationship between reversible fouling rate and flux, whilst reversible fouling rate increased exponentially with turbidity. The increased fouling rate deceased the period between CIPs, and was associated with the transformation of reversible fouling to irreversible fouling.

Extrapolation of the fouling rate:turbidity relationship to zero turbidity revealed an exponential relationship between reversible fouling rate and flux in the absence of measurable turbidity. It can be concluded that other water quality determinants contributed to both reversible and irreversible fouling, with the proportional contribution to reversible fouling increasing with decreasing turbidity.

Results indicate that, for full-scale plant, the sustainable flux is a more representative and useful parameter than the critical flux, since the latter does not take into account the process economics.

5.5 References

Amin, I.N.H.M., Mohammad, A.W., Markom, M., Peng, L.C., Hilal, N. (2010), Flux decline study during ultrafiltration of glycerine-rich fatty acid solutions, *Journal of Membrane Science*, 351, p. 75-86.

Bacchin, P. (2004), A possible link between critical and limiting flux for colloidal systems: consideration of critical deposit formation along a membrane, *Journal of Membrane Science*, 228, p. 237-241.

Bacchin, P., Aimar, P. (2005), Critical fouling conditions induced by colloidal surface interaction: from causes to consequences, *Desalination*, 175, p. 21-27.

Bacchin, P., Aimar, P., Field, R.W. (2006), Critical and sustainable fluxes: Theory, experiments and applications, *Journal of Membrane Science*, 281, p. 42-69.

Bessiere, Y., Abidine, N., Bacchin, P. (2005), Low fouling condition in dead-end filtration: Evidence for a critical filtered volume and interpretation using critical osmotic pressure, *Journal of Membrane Science*, 264, p. 37-47.

Bourgeous, K.N., Darby, J.L., Tchobanoglous, G. (2001), Ultrafiltration of wastewater: effect of particles, mode of operation, and backwash effectiveness, *Water Research*, 35 (1), p. 77-90.

Brookes, A., Jefferson, B., Guglielmi, G., Judd, S.J. (2006), Sustainable flux fouling in a membrane bioreactor: Impact of flux and MLSS, *Separation Science and Technology*, 41, p. 1279-1291.

Carić, M.D., Milanović, S.D., Krstić, D.M., Tekić, M.N. (2000), Fouling of inorganic membranes by adsorption of whey proteins, *Journal of Membrane Science*, 135, p. 83-88.

Cho, J., Amy, G., Pellegrino, J. (2000), Membrane filtration of natural organic matter: factors and mechanisms affecting rejection and flux decline with charged ultrafiltration (UF) membrane, *Journal of Membrane Science*, 164, p. 89-110.

Citulski, J.A., Farahbakhsh, K., Kent, F.C. (2009), Effects of total suspended solids loading on short-term fouling in the treatment of secondary effluent by an immersed ultrafiltration pilot system, *Water Environment Research*, 81, p. 2427-2436.

Hwang, K.-J., Chan, C.-S., Tung, K.-L. (2009), Effect of backwash on the performance of submerged membrane filtration, *Journal of Membrane Science*, 330, p. 349-356.

Jarsutthirak, C., Amy, G., Croué, J-P. (2002), Fouling characteristics of wastewater effluent organic matter (EfOM) isolates on NF and UF membranes, *Desalination*, 145, p. 247-255.

Jiang, T., Kennedy, M.D., Guinzbourg, B.F., Vanrolleghem, P.A., Schippers, J.C. (2005), Optimising the operation of a MBR pilot plant by quantitative analysis of the membrane fouling mechanism, *Water Science and Technology*, 51 (6-7), p. 19-25.

Lazarova, V., Gallego, S., García Molina, V., Rougé, P. (2008), Problems of operation and main reasons for failue of membranes in tertiary treatment systems, *Water Science and Technology*, 57 (11), p. 1777-1784.

Le-Clech, P., Jefferson, B., Chang, I.S., Judd, S.J. (2003), Critical flux determination by the flux-step method in a submerged membrane bioreactor, *Journal of Membrane Science*, 227, p. 81-93.

Le-Clech, P., Chen, V., Fane, T.A.G. (2006), Fouling in membrane bioreactors used in wastewater treatment, *Journal of Membrane Science*, 284, p. 17-53.

Lee, N., G. Amy, J-P. Croué J-P., Buisson, H. (2004), Identification and understanding of fouling in low-pressure membrane (MF/UF) filtration by natural organic matter (NOM), *Water Research*, 38, p. 4511-4523.

Li, Y., Zhang, W., Zhang, X., Chen, C., Wang, J. (2010), Characterisation of fouling in immersed polyvinylidene fluoride hollow fibre membrane ultrafiltration

by particles and natural organic matter, *Desalination and Water Treatment*, 18, p. 309-314.

Lin, H., Bérubé, P.R. (2007), Modelling the impact of permeate flux and hydrodynamic conditions on fouling in submerged hollow fibre membranes, Water Science and Technology: Water supply, 7 (4), p. 111-118.

Liu, Q-F., Kim, S-H., Lee, S. (2009), Prediction of microfiltration membrane fouling using artificial neural network models, *Separation and Purification Technology*, 70, p. 96-102.

McAdam, E.J., Judd, S.J. (2008), Optimisation of dead-end filtration for an immersed anoxic membrane bioreactor, *Journal of Membrane Science*, 325, p. 940-946.

Mousa, H.A., Al-Hitmi, S.A. (2007), Treatability of wastewater and membrane fouling. *Desalination*, 217, p. 65-73.

Nguyen, T., Fan, L., Roddick, F.A., Harris, J.L. (2009), A comparative study of microfiltration and ultrafiltration of activated sludge-lagoon effluent, *Desalination*, 236, p. 208-215.

Ognier, S., Wisniewski, C., Grasmick, A., Membrane fouling during constant flux filtration in membrane bioreactors, *Membrane Technology July 2002*.

Parameshwaran, K., Fane, A.G., Cho, B.D., Kim, K.J. (2001), Analysis of microfiltration performance with constant flux processing of secondary effluent, Water Research, 35 (18), p. 4349-4358.

Psoch, C., Shiewer S. (2006), Resistance analysis for enhanced wastewater membrane filtration, *Journal of Membrane Science*, 280, p. 284-297.

Raffin, M., Germain, E., Judd, S. (2011), Optimising operation of an integrated membrane system (IMS) - A Box-Behnken approach, *Desalination*, 273, p. 136-141.

She, Q., Tang, C.Y., Wang, Y-N., Zhang, Z. (2009), The role of hydrodynamic conditions and solution chemistry on protein fouling during ultrafiltration, *Desalination*, 249, p. 1079-1087.

US EPA, Membrane Filtration guidance manual, EPA 815-R-06-009, November 2005.

Wang, S-S. (1988), Effect of solution viscosity on ultrafiltration flux, *Journal of Membrane Science*, 39, p. 187-194.

Wang, L., Wang, X., Fukushi, K-I. (2008), Effects of operational conditions on ultrafiltration membrane fouling, *Desalination*, 229, p. 181-191.

Zheng, X., Ernst, M., Jekel, M. (2009), Identification and quantification of major organic foulants in treated domestic wastewater affecting filterability in dead-end filtration, *Water Research*, 43, p. 238-244.

CHAPTER 6: OPTIMISATION OF MF MEMBRANE CLEANING PROTOCOL IN AN INDIRECT POTABLE REUSE (IPR) SCHEME

Raffin, M., Germain, E., Judd, S. (2011), Optimisation of MF membrane cleaning protocol in an Indirect Potable Reuse (IPR) scheme, *Separation and Purification Technology*, 80, 452-458.

6.1 Introduction

Microfiltration (MF) and ultrafiltration (UF) membranes are widely used for pre-treatment of reverse osmosis (RO) processes in wastewater recovery for indirect potable reuse (IPR). However, fouling of the MF/UF membranes dramatically reduces the throughput. Reversible fouling is readily controlled by periodic backwashing, whereas irreversible fouling requires chemical cleaning. The extent of irreversible fouling strongly depends on feedwater quality, which then influences the chemical cleaning protocol needed (or cleaning in place, CIP).

Many studies of MF/UF membrane CIP protocols have been reported, and recently reviewed (Porcelli and Judd, 2010a). However, relatively few have been based on municipal wastewater, notwithstanding the large number of publications on membrane fouling. Optimisation of membrane CIP is often performed on an ad hoc basis during routine maintenance of pilot (Brant et al, 2010; Bartels *et al.*, 2004) or full-scale plants (Lazarova *et al.*, 2008). This is inevitably time-consuming, since the membranes must become irreversibly fouled prior to each measurement of CIP efficacy.

Laboratory-scale study offers an alternative for cleaning optimisation. Most laboratory-scale studies, however, rely on samples of virgin membrane to allow experiments to be conducted under the same conditions (Tian *et al.*, 2010), and also often use either surrogate foulants (Bartlett *et al.*, 1995; Väisänen *et al.*, 2002) or sampled natural water (Zondervan and Roffel, 2007) of constant water quality - including feed water temperature (Mendoza-Roca *et al.*, 2010; Popocic *et al.*, 2009). As such, actual fouling conditions of an operational plant, which are subject to wide diurnal and seasonal fluctuations in feedwater quality, are not necessarily well represented. Finally, whilst some studies have incorporated multiple filtration cycles and constant flux (Zondervan and Roffel, 2007; Chen *et al.*, 2003), most have employed constant pressure and/or a single filtration cycle

only (Tran-Ha and Wiley, 1998; Yu et al, 2010) which precludes extrapolation of experimental data to full scale operation (Heijman *et al.*, 2007).

Only four published laboratory-scale studies have used aged membranes, one using membranes fouled at bench scale (Tang *et al.*, 2010). The study of Strugholtz *et al.* (2005) compared different cleaning reagents at different temperatures, concentrations and different cleaning sequences, providing semi-quantitative data for permeability recovery based on a single fouled capillary for each experiment. Porcelli *et al.* (Porcelli *et al.*, 2009; Porcelli and Judd, 2010b) employed membrane modules fabricated from membrane hollow fibre (HF) fouled through full-scale application, and found fouling to become more tenacious with membrane age. This was also demonstrated by Tang *et al.* (2010), who found denser biofilms formed on used membranes compared with virgin material. Whilst demonstrating the efficacy of the experimental method, Porcelli *et al.* were not able to demonstrate a single optimum cleaning protocol for a specific installation since only a limited range of reagents were trialled for each site reported.

This work aimed to optimise an MF membrane CIP using membrane fibres taken from a single pilot plant treating secondary municipal wastewater, with results from the lab-scale study being compared with the CIP performance recorded at pilot scale. The influence of filtration temperature on water permeability recovery was assessed, and a cost benefit analysis performed to determine optimum CIP conditions with respect to energy, chemical consumption and net water production. Box-Behnken design was used to statistically determine optimum CIP parameters for the MF unit, as previously reported (Porcelli *et al.*, 2009; Porcelli and Judd, 2010b).

6.2 Materials and methods

6.2.1 Pilot plant overview

The 600 $m^3.d^{-1}$ Indirect Potable Reuse (IPR) pilot plant has been described elsewhere (Raffin *et al.*, 2011; Annex 3). Final effluent from a conventional activated sludge plant passes through a pre-filter (*Bollfilter*) before being filtered by a submerged microfiltration (MF) unit (Memcor). The MF permeate then passes through a reverse osmosis (RO) system (Hydranautics) and on to an advanced oxidation process (AOP) and a degasser tower before undergoing pH correction. Chloramine can be dosed at three different points in the process, pre pre-filter, pre MF and pre RO, to control biofouling. The plant is fully automated and data recorded on a supervisory control and data acquisition (SCADA) system.

6.2.2 Bench-scale permeability test rig

To optimise the cleaning of the MF membranes, a bench-scale permeability test rig was used, identical to that first reported by Porcelli *et al.* (2009). The simple apparatus comprises a constant head of 2 m water applied to a membrane module constructed from lengths of HF membrane samples. The same apparatus was used for testing the CIP efficacy on 12 fouled HF filaments sampled from a single MF module extracted from the pilot plant. To limit heterogeneity of the bench-scale module, all the fibres were extracted within 25 mm central region of the module.

Membrane permeability K ($l.m^{-2}.h.bar^{-1}$) is the ratio of the permeate flux J ($l.m^{-2}.h$, or LMH) to the trans-membrane pressure (TMP, bar), normalised to 20°C:

$$K = \frac{J}{TMP} \times 1.6124 \times e^{(-0.0238 \times T)} \quad \textbf{(6-1)}$$

where T is the permeate temperature in °C. The %permeability recovery (%R_{exp}) from the CIP is then given by:

$$\%R_{\exp} = \frac{K_f - K_i}{K_i} \times 100 \quad \textbf{(6-2)}$$

where K_i and K_f is the permeability before and after the CIP.

6.2.3 MF membrane module

The commercial hollow fibre (HF) membrane studied was the Memcor *S10V* microfilter (Table 6-1). The membrane module was removed from the MF unit after two weeks of operation at 40 LMH and 30 min filtration cycle. During these two weeks, an average turbidity of 3.25 NTU ± 52% was observed as well as an average Total organic carbon (TOC) concentration of 5.88 $mg.L^{-1}$ ± 5.1%. Average temperature, conductivity and pH were respectively 22°C ± 3.4%, 1130 $\mu S.cm^{-1}$ ± 2.1% and 7 ± 0.9%, and the chloramine dose was maintained constant at 1 $mg.L^{-1}$ during the entire period. A backwash was performed before the module was extracted from the MF unit to remove reversible fouling.

Table 6-1 Membrane module specifications

Manufacturer	Siemens Water Technologies Memcor Ltd
Membrane Type	XS CMF-S S10V
Materials	PVDF
Area/module	25.3 m^2
Configuration	1 cell of 16 submerged hollow fibre modules (with possible expansion to 24)
Filtration mode	Out-in
Backwash	Air + water
Operating temperature	>0-40ºC (max 45ºC)
Operating pH	2-10.5
Standard CIP	600L NaOCl solution (540 ppm, 30ºC) followed by 600L H_2SO_4 solution (pH 2.5, 30ºC)

6.2.4 Cleaning protocol

Four parameters were identified as potentially affecting CIP efficacy: chemical type, chemical concentration (C, ppm or w/w% or pH), soaking time (S, min) and cleaning solution temperature (T, °C). Table 6-2 provides a summary of the different parameters for each chemical studied. To assess the impact of basicity, additional experiments were performed using NaOH alone at pH 10,

varying temperature and soak time from 10 to 40^{o}C and 60 to 120 minutes respectively. Chemical reagents were introduced into the hollow fibres of the bench module to displace all process water, and the module placed in a 250 ml container filled with cleaning solution placed in an isothermal bath for the required soak period.

6.2.5 Experimental design

Box-Behnken design (Meyers *et al.*, 2009) was used to fit a second-order response surface to determine optimum CIP conditions. Besides providing a robust design, this method allows the number of experiments to be reduced from 27 to 15 for a 3^{n} factorial design by studying the influence of parameters at three equidistant values; the combinations of values for the three parameters can be found in Raffin *et al.* (2011). For NaOH at pH 10 A 3^{n} factorial design was performed since only two parameters were assessed in this case.

Table 6-2 Parameters and their value ranges

Parameter #		Oxidising reagent		
		NaOCl [**,***]	***NaOCl (pH=10)*** [**]	***H_2O_2*** [**,****]
	Parameters		***Range***	
x_1	C (ppm or w/w%)	300-900	300-900	0.5-1.5 %
x_2	S (min)	60-120	60-120	60-120
x_3	T (ºC)	10-40	10-40	10-40
		Acidic reagent		
		H_2SO_4 [*]	***HCl*** [*]	***Citric acid*** [*]
	Parameters		***Range***	
x_1	C (pH)	2-4	2-4	2-4
x_2	S (min)	15-45	15-45	15-45
x_3	T (^{o}C)	10-40	10-40	10-40

[*]Experiments performed at an average permeate temperature of 30ºC during permeability test
[**]Experiments performed at an average permeate temperature of 10ºC during permeability test
[***]pH varying from 9.25 (300 ppm) to 9.90 (900 ppm) as a function of NaOCl concentration
[****]pH varying from 4.5 (1.5 w/w%) to 5.4 (0.5 w/w%) as a function of H_2O_2 concentration

Multiple linear regression was used to generate the model equation:

$$\%R_{predicted} = \beta_0 + \sum_{i=1}^{3} \beta_i x_i + \sum_{i=1}^{3} \beta_{ii} x_i^2 + \sum \sum_{i<j=2}^{3} \beta_{ij} x_i x_j \quad \textbf{(6-3)}$$

where $\%R_{predicted}$ is the predicted permeability recovery (%), β_0 the intercept coefficient, β_i the coefficients in the polynomial function, β_{ii} the quadratic coefficients, β_{ij} the interaction coefficient and x_i and x_j the factors to be studied, where *i* takes values of 1 to 3 and *j* values of 2 to 3. An analysis of variance (ANOVA) table was produced for each regression to check if the model equations reflected the experimental data.

6.2.6 Supplementary tests

The cleaning solutions used for each experiment were analysed for total and organic carbon (TOC and DOC respectively), specific UV absorbance and 30 inorganic ions. Membrane autopsies were performed according to previously reported methods (Porcelli *et al.*,2009), whereby 2.5 x 10^{-3} m^2 of membrane were blended in 500 ml of deionised water for 10 minutes to dissolved inorganic/organic membrane foulants. The eluates were then filtered through a 0.45 µm filter and analysed.

6.3 Results and discussion

6.3.1 Permeability recovery

Permeability recovery determined from each experiment was found to vary from -12% for citric acid to 142% for NaOCl at pH 10 (Table 6-3). Results indicated oxidising reagents to be the most efficient reagents for permeability recovery, followed by caustic and then acidic reagents. Similar trends were observed in operating the pilot plant, where an average recovery of ~50% had been recorded following the NaOCl clean with negligible permeability recovery after applying H_2SO_4, and also corroborate previous reported data from a chemical study based on ten cleaning reagents (Zondervan and Roffel, 2007). The variance data shows variability to be higher for oxidising agents, such that the conditions applied (concentration, temperature and soaking time) have more influence on oxidising reagents than on acidic reagents.

NaOH permeability recovery efficiency was lower than that for oxidising reagents, in agreement with findings from some studies (Bohner and Bradley, 1990) but contrary to those from other studies (Zondervan and Roffel, 2007; Strugholtz *et al.*, 2005) where more highly alkaline solutions than those employed in the current study were studied. In these latter cases, the PES membranes used were not constrained by alkaline hydrolysis as is the case PVDF membranes.

Regression analysis was applied to each set of experiments to determine the second-order model equation for each cleaning reagent. Table 6-4 provides the linear (C, T, S), quadratic (T^2, C^2, S^2) and interaction (TC, TS, CS) coefficients of Equation (6-3) as well as the coefficient of determination (R^2). R^2 values reveal the experimental data to fit well with the modelled data for NaOCl at pH 10, NaOCl at unadjusted pH, HCl and citric acid. Apart for NaOH at pH 10, regression equations were highly statistically significant (p-value< 0.05 in the ANOVA table) for all cleaning reagents, although lower R^2 values were obtained for H_2O_2 and H_2SO_4 associated with more highly scattered data. The lower significance of the regression for NaOH probably reflects the lower number of experiments performed (9 instead of 15 for the other chemicals) due to the necessary omission of reagent concentration.

From the model equations, the maximum achievable permeability recovery and the associated conditions were defined (Table 6-5). NaOCl buffered at pH 10 was found to be the only cleaning reagent providing a permeability recovery exceeding 100%. The model equations were used to compute the temperature as a function of the NaOCl concentration required to attain 100% recovery at three different soak times (Figure 6-1). The figure indicates a trade-off between cleaning reagent temperature and concentration, and process downtime.

Table 6-3 Permeability recovery (mean, minimum, maximum and variance) for each chemical reagent

Chemical reagent	Average %R	Max %R	Min %R	Variance
NaOCl at pH 10	52	142	7.0	1698
NaOCl	41	86	-5.0	864
H_2O_2	16	96	-2.8	689
NaOH at pH 10	12	40	-10	282
H_2SO_4	3.4	33	-4.0	86
HCl	3.4	16	-3.6	36
Citric acid*	5.2	19	-12	90

Table 6-4 Second order model coefficient (β, Equation 3) and coefficient of determination R^2 for each chemical reagent.

Reagent	Intercept	T	C	S	T^2	C^2	S^2	TC	TS	CS	R^2
NaOCl pH 10	61.5	-2.88	0.07	-1.53	0.07	0.00	0.01	0.00	0.02	0.00	0.97
NaOCl	86.3	0.67	0.01	-2.22	0.02	0.00	0.01	0.00	0.01	0.00	0.99
H_2O_2	118	-2.45	-30.1	-2.09	0.01	49.4	0.02	0.82	0.02	-1.25	0.78
NaOH pH 10	-93.7	5.91	-	0.68	-0.01	-	-0.00	-	-0.00	-	0.87
HCl	-28.8	-0.68	24.7	-0.23	0.01	-3.91	0.00	0.08	0.02	0.01	0.97
H_2SO_4	44.1	-0.91	-38.2	1.68	0.04	7.68	-0.02	-0.31	0.00	-0.13	0.76
Citric acid	116	-0.70	-39.8	-3.11	0.00	4.99	0.02	0.04	0.03	0.20	0.91

Table 6-5 Maximum achievable permeability recovery predicted by the model equations and conditions required for each chemical reagent studied

Cleaning reagent	Max $\%R_{predicted}$	Conditions to fill to achieve max %R		
		Temperature	***Concentration***	***Soaking time***
NaOCl pH = 10	>100%	See Figure 2		
NaOCl	97%	40ºC	900 ppm	60 min
H_2O_2	91%	40ºC	0.5 w/w%	120 min
NaOH pH 10	33%	28^{o}C	-	120 min
HCl	17%	40ºC	pH 3.6	120 min
H_2SO_4	25%	40ºC	pH 2	32 min
Citric acid	20%	40ºC	pH 2	15 min

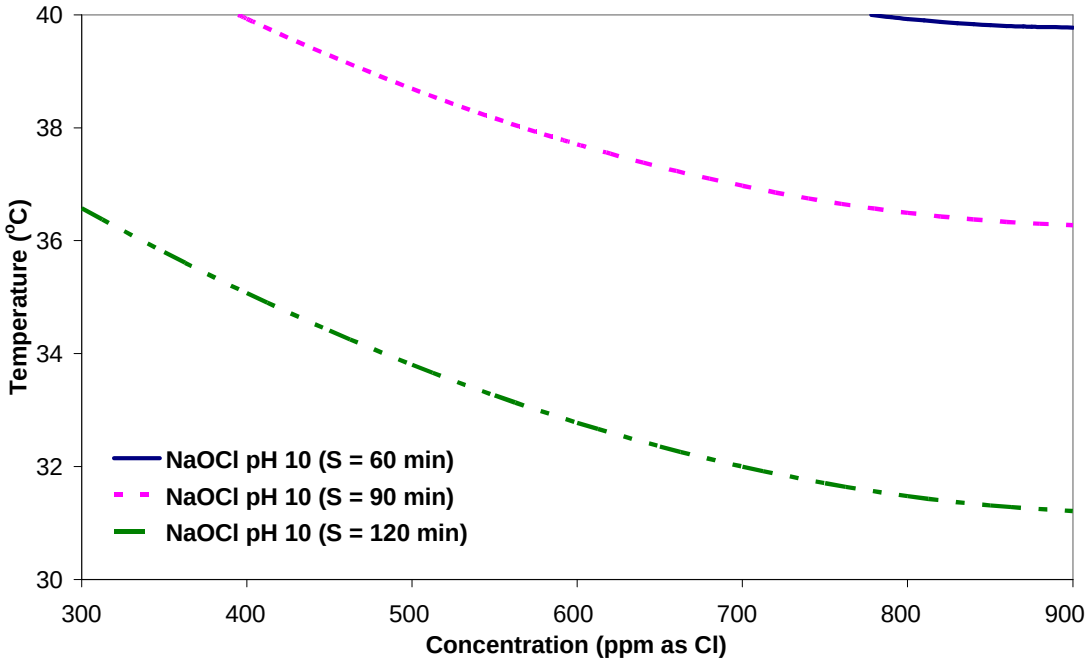


Figure 6-1 Temperature and concentration combination required to reach 100 % permeability recovery for different soak times.

Figure 6-2 to Figure 6-7 show the general effects of each parameter on permeability recovery for each cleaning reagent. Figure 6-2and Figure 6-3 indicates an expected benefit from increasing temperature for most cleaning reagents, with a maximum recovery at 28^{o}C for NaOH at pH 10. For H_2SO_4 there is a minimum negative recovery at ~25ºC, but recoveries are low (-4 to 8%). It is generally agreed that cleaning efficiency increases with temperature since it enhances reaction kinetics and foulant solubility (Chen *et al.*, 2003; Porcelli *et al.*, 2009; Li *et al.*, 2005). However, other studies have shown that an optimum temperature exists after which the permeability recovery decreases (Bartlett *et al.*, 1995). The influence of concentration (Figure 6-4and Figure 6-5) varies between oxidative and acidic reagents, with the expected positive benefit for NaOCl at both pH 10 and uncorrected pH (Figure 6-4), but only at concentrations above ~500 mg/l as Cl in the case of the latter. This is readily attributed to the impact of pH, since the NaOCl solution is alkaline. Figure 6-5

also shows that permeability recovery decreases with H_2O_2 concentration up to ~1.1 wt%, likely to be due to the decreasing pH with increasing peroxide concentration (from 5.4 at 0.5 wt% to 4.5 at 1.5 wt%) such as any minor benefit from an increased oxidation potential is countered by the negative influence of the decreasing pH . The impact of soak time (Figure 6-6 and Figure 6-7) also varies, with the expected positive effect for H_2O_2, NaOCl at pH 10 and NaOH at pH 10, but a small negative impact for unadjusted NaOCl (Figure 6-6), which again is associated with a decrease in pH with soak time due to the reductive degradation of hypochlorite to acidic products. For acidic reagents (Figure 6-7), increasing soak time provides no additional benefit and, indeed, tends to reduce permeability recovery.

Negative permeability recoveries, indicating an increased fouling layer resistance following CIP, were found for oxidising, alkali and acidic reagents, corroborating previous findings (Väisänen *et al.*, 2002; Mohammadi *et al.*, 2003). This negative effect is generally attributed to a change in the foulant layer morphology on contact with the different chemical reagents. Extreme temperature and pH can lead to precipitation, solidification and gel formation or swelling of the fouling layer leading to a decrease in voidage in the fouling layer (Bartlett *et al.*, 1995). Although no previous studies reported negative permeability recovery with H_2O_2 or NaOCl, the current study suggests that low cleaning temperatures (<~10ºC) may produce negative permeability recoveries even for alkaline hypochlorite. Permeability recoveries above 100% are a consequence of the index used to determine the permeability recovery (Eq. 6-2), where the change in permeability on cleaning may exceed the initial membrane permeability if the membrane is heavily fouled and the reagent particularly effective. It is also possible for membrane deterioration through alkaline hydrolysis to take place for the PVDF material, contributing to the decreased membrane resistance, though this would be expected to be minimal at the mildly alkaline pH employed.

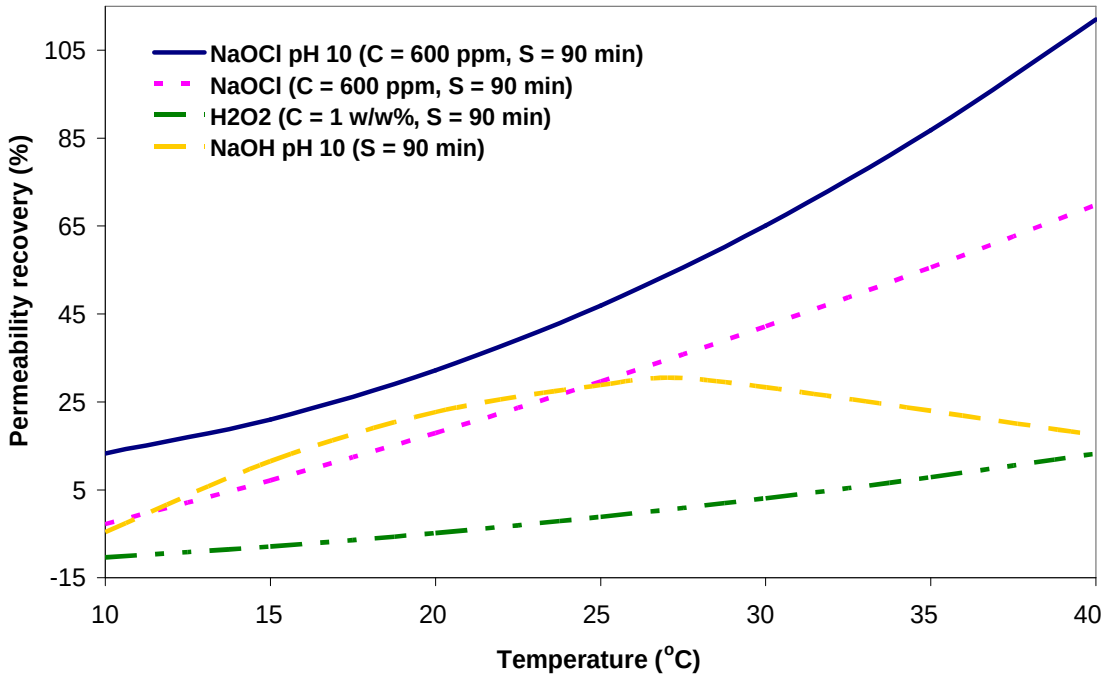


Figure 6-2 Permeability recovery (%) as a function of temperature for oxidising reagents

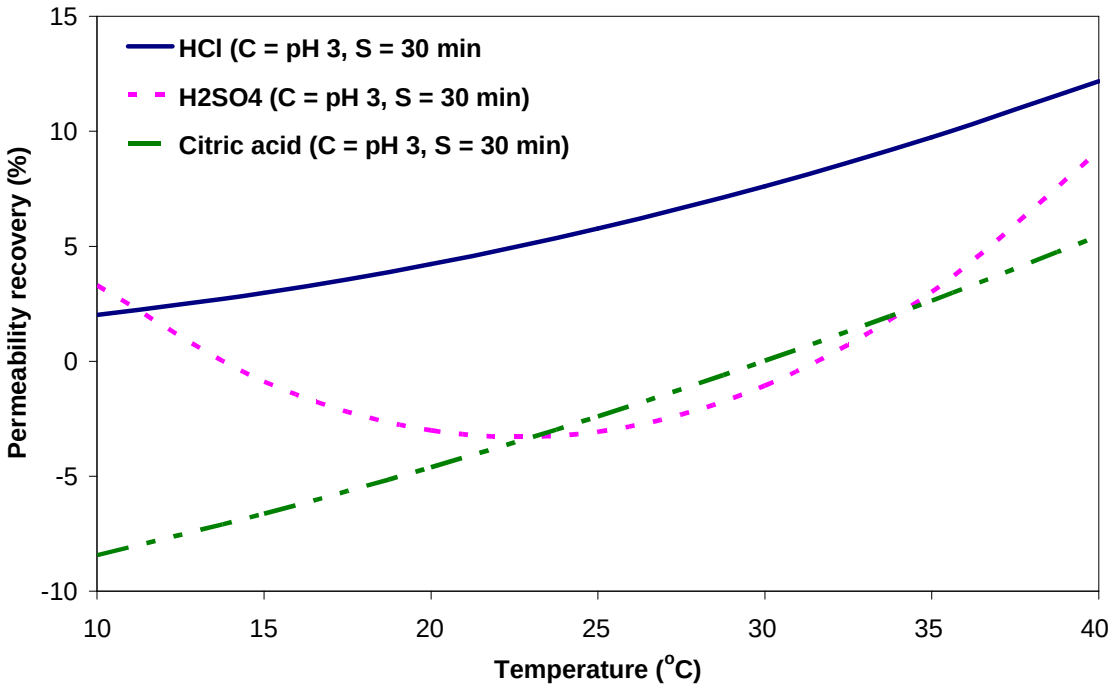


Figure 6-3 Permeability recovery (%) as a function of temperature for acidic reagents

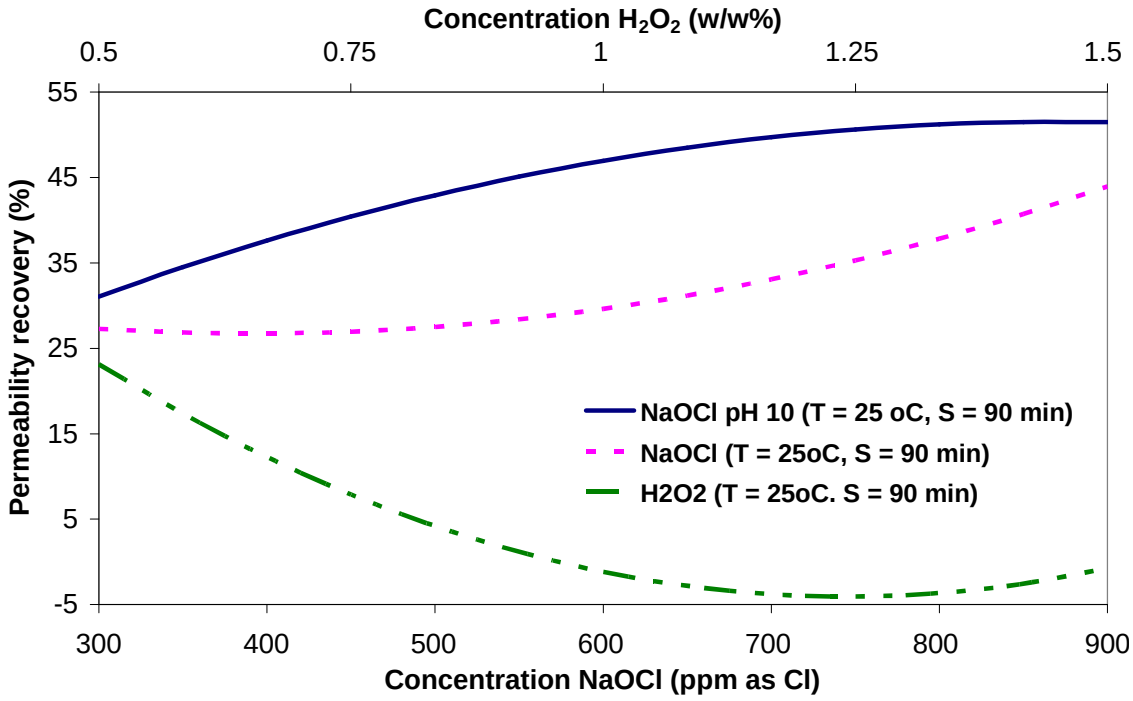


Figure 6-4 Permeability recovery (%) as a function of concentration for oxidising reagents

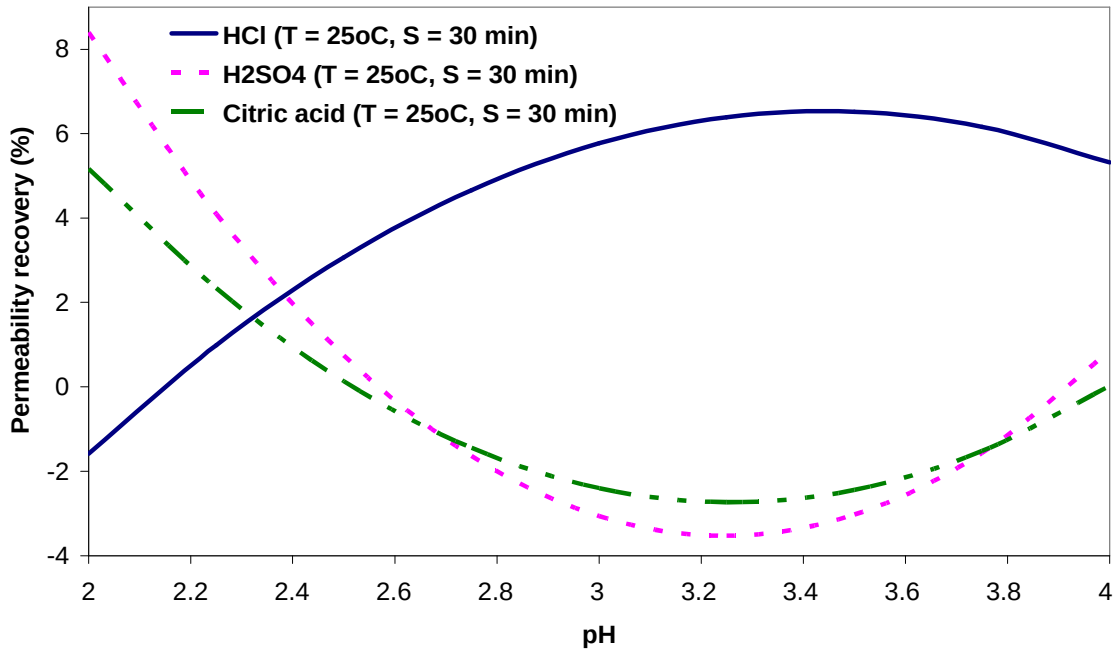


Figure 6-5 Permeability recovery (%) as a function of pH for acidic reagents

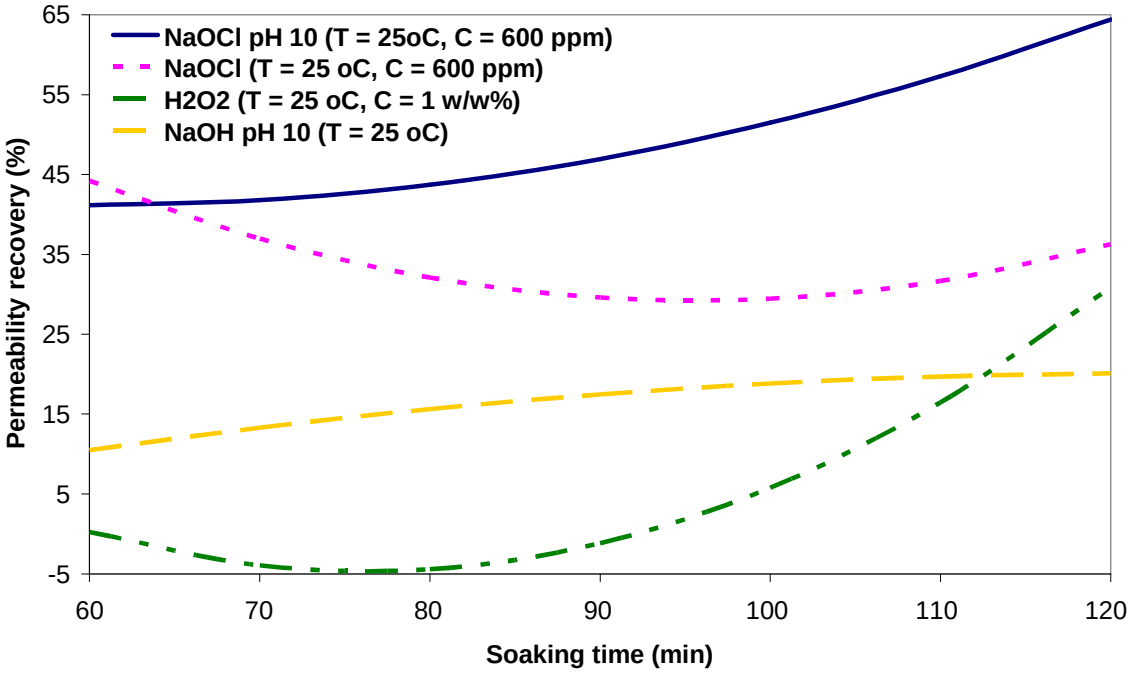


Figure 6-6 Permeability recovery (%) as a function of soak time for oxidising reagents

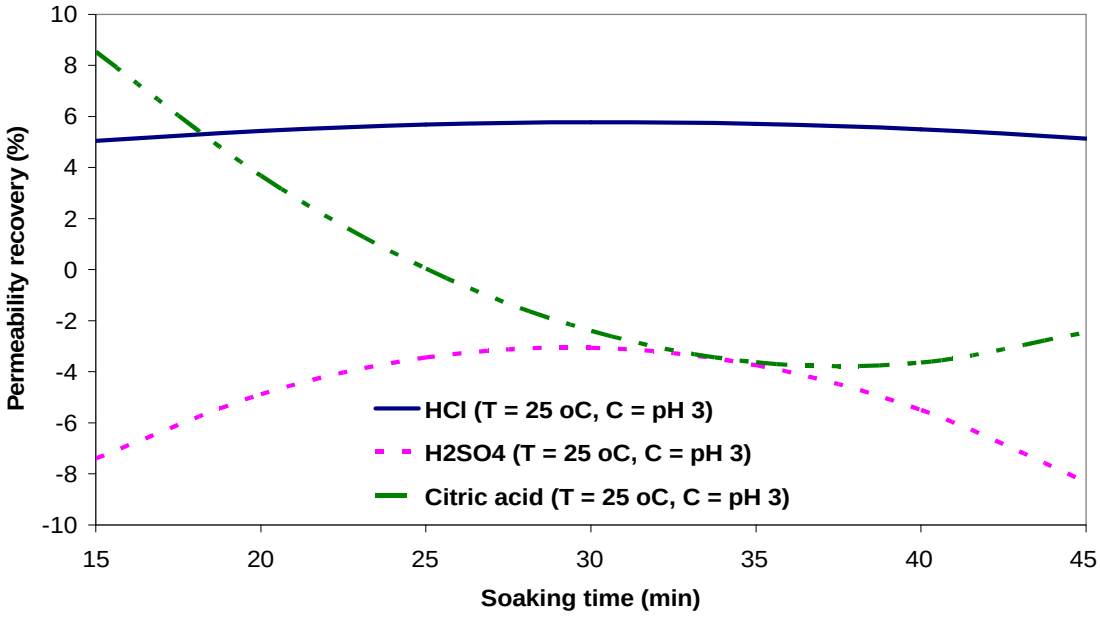


Figure 6-7 Permeability recovery (%) as a function of soak time for acidic reagents

Analysis of the p-value (from the ANOVA table) for each coefficient shown in Table 6-4 provides an indication of those parameters having a significant impact (p-value < 0.05) on permeability recovery for each cleaning reagent (Table 6-6). Data indicate that the influence of the three parameters varies across the five reagents studied. As shown in Table 6-6, quadratic and interaction coefficients are highly significant for all cleaning conditions; synergistic effects are evidently important in chemical cleaning.

Table 6-6 Significance of parameters for each chemical reagent

Cleaning reagents	**Coefficient with a p-value < 0.05**	**Parameters to assess**		
		Temperature	*Concentration/pH*	*Soak time*
NaOCl pH 10	T^2, TS, C	✓	✓	✓
NaOCl	T, S, S^2, C^2, CS	✓	✓	✓
H_2O_2	S^2, CS, C^2		✓	✓
NaOH pH 10	T, T^2	✓	N/A	
HCl	T^2, C, C^2	✓	✓	
H_2SO_4	T^2, TC, C^2	✓	✓	
Citric acid	S, TS	✓		✓

N/A not applicable

6.3.2 Water quality

Water quality analysis of the feed and permeate water showed the MF process to remove suspended solids, microbiological compounds, some organic compounds (as total organic carbon) and some inorganic compounds (such as aluminium and iron). It is thus expected for these compounds to irreversibly foul the membrane, not withstanding their substantial removal by the physical cleaning cycle.

Autopsy of fouled and cleaned membrane fibres revealed that, of the detectable compounds, TOC was highest in concentration (up to 300 mg/m^2 of membrane). Of the 30 elements analysed, only five were consistently detected in the eluates following CIP: barium (up to 35 mg/m^2 of membrane), boron (up

to 25 mg/m^2), zinc (up to 0.7 mg/m^2), iron (up to 2.8 mg/m^2) and aluminium (up to 3.0 mg/m^2), indicating irrecoverable fouling by these species. Citric acid was the only reagent for which no iron was detected in the membrane eluates, reflecting the known complexation proclivity of citric acid with iron in particular (Porcelli and Judd, 2010a). Foulants were thus not completely removed even at permeability recoveries above 100% (e.g. for NaOCl, pH 10) – an observation made by previous workers (Bohner and Bradley, 1990) who suggested that full permeability recovery results from channels through the filter cake created by cleaning and rinsing. It may also indicate that oxidising and acidic reagents must both be applied to maximise permeability recovery.

Cleaning solution eluates were also chemically analysed before and after cleaning, and the average removal in mg element per square meter of membrane calculated (Table 6-7). As expected, and in accordance with Strugholz *et al.* (2005), NaOCl both at corrected and non-corrected pH was generally more efficient at removing organic compounds and acidic agents more effective against inorganic matter such as boron, manganese, iron and tin – but comparable with NaOCl for removing hardness ions. Aluminium was better removed at higher pH (NaOH pH 10), reflecting its amphoteric nature and contrary to previous reports where citric acid was found to be most efficacious (Strugholtz *et al.*, 2005). These authors found the alkaline reagents NaOH and alkaline oxidant to perform better than inorganic acid, in agreement with the current study. NaOH at pH 10 and H_2O_2, whilst less effective against organic matter than other oxidising agents, yielded better inorganic matter removal than the other oxidising reagents, also corroborating previous reports (Strugholtz *et al.*, 2005). Contrary to expectation, TOC removed by H_2O_2 was lower than for any of the other reagents and TOC removal for NaOH was similar to that attained by acidic reagents.

For both autopsy and cleaning eluates, the specific UV absorbance (SUVA) was below 2 for all the tests, suggesting that organic foulants removed during cleaning were mostly hydrophilic - in accordance with previous similar studies of NOM fouling of PVDF membranes (Kimura *et al.*, 2006). Other studies have

also identified NaOCl as the best cleaning reagent for removal of hydrophilic organic compounds (Zondervan an Roffel, 2007), reporting a reduced efficiency for hydrophobic and inorganic compounds. NaOCl at high pH is thought to allow hydrophobic and hydrophilic fractions of the organic matter to be eluted, with the NaOH hydrolysing organic compounds to create a more porous fouling layer to expedite the action of the oxidant (Strugholtz *et al.*, 2005). The SUVA of the MF feed water was also below 2. Statistical analyses performed on both autopsy and cleaning eluates showed no clear trends relative to the experimental conditions or permeability recoveries, possibly reflecting the heterogeneous nature of membrane fouling.

Table 6-7 Average compounds removal (mg per square meter of membrane) for each cleaning reagent

Element	*NaOCl*	*NaOCl pH 10*	*NaOH pH 10*	*H_2O_2*	*Citric acid*	*H_2SO_4*	*HCl*
	Average compounds removal (mg per square meter of membrane)						
Boron	1.47 ± 0.09	1.57 ± 0.05	1.74 ± 0.07	1.76 ± 0.08	1.97 ± 0.96	2.38 ±0.22	3.01 ± 1.51
Calcium	17.6 ± 2.64	6.41 ± 8.55	18.98 ± 2.75	0.00 ± 0.00	19.3 ± 37.0	3.13 ± 7.05	23.4 ± 7.05
Manganese				0.07 ± 0.03	0.02 ± 0.04	0.01 ± 0.1	0.02 ± 0.04
Iron	0.13 ± 0.28	0.17 ± 0.13	0.21 ± 0.09	0.25 ± 0.16	0.59 ± 0.76	0.57 ± 1.4	0.25 ± 0.66
Aluminium	0.76 ± 1.00	0.21 ± 0.53	1.10 ± 0.69	0.23 ± 0.08	0.15 ± 0.43	0.13 ± 0.77	0.34 ± 0.77
Tin				0.06 ± 0.03	0.81 ± 1.17	0.43 ± 1.33	0.01 ± 0.02
TOC	55.3 ± 30.6	50.1 ± 28.0	21.0 ± 9.67	3.53 ± 1.44	N/A	14.8 ± 38.2	23.2 ± 38.1

A comparison of permeability recovery (Table 6-3) and compounds removal (Table 6-7) indicates that whilst compounds removal for both pH-corrected and uncorrected NaOCl are similar, the permeability recovery for the former is higher than that for the latter. This suggests that the alkaline pH assists the oxidative action of the NaOCl through swelling of the membrane and/or fouling layer, creating a more open structure and leading to greater permeability recovery. NaOH at pH 10 alone removed less than 40% of the organic matter

removed by the hypochlorite solutions, similar in performance to the mineral acids, but with the latter providing negligible permeability recovery, as reported previously (Zondervan and Roffel, 2007; Strughlotz *et al.*, 2005), compared to the mildly alkaline solution. H_2O_2 provided negligible removal of compounds, including TOC, but nonetheless provided a slightly higher permeability recovery than the dilute NaOH.

6.3.3 Cost

Whilst NaOCl at pH 10 provided the highest recovery, different combinations of temperature, concentration and soak time can be applied to attain 100% recovery (Figure 6-1). Table 8 indicates the costs involved per megalitre (ML) of water produced at different soak times for different temperatures and NaOCl concentrations. This cost includes the chemical usage (NaOCl and NaOH) and the energy consumption required for heating (Table 6-8) calculated for a flow of 20 m^3/h, a backwash frequency of 30 minutes and CIP frequency of 15 days. The CIP cost per ML of permeate decreases most significantly with soak time, since for the same chemical usage the energy consumption required for the heating system is lower at longer soak periods (Table 6-8); heating energy costs representing 75% to 86% of the total cost. A slight decrease in cost arises with increasing NaOCl concentration, since this permits a lower temperature to achieve the same recovery (Figure 6-2).

Table 6-8 Cost values and outputs

Parameter		Value
Cost factor		
Energy consumption (GBP/kWh)	CIP tank heating system:	0.17*
Chemicals (GBP/kg)	NaOCl (14-15 %):	0.30
	NaOH (35%):	3.47
Outputs: Cost, GBP/ML permeate for 100% permeability recovery at		
S = 60 minutes, 780 – 900 mg/L NaOCl		1.56 – 1.58
S = 90 minutes, 400 – 900 mg/L NaOCl		1.39 – 1.46
S = 120 minutes, 300 – 900 mg/L NaOCl		1.14 – 1.26

*Porcelli and Judd (2010c)

6.3.4 Methods validation

The effect of water filtration temperature on permeability results was evaluated by duplicating the test using NaOCl at filtration temperatures of 10^{o}C and 30^{o}C on the bench scale (Table 6-9). It appears that even after temperature-correcting permeability to 20^{o}C, results for the same chemical cleaning at different filtration temperatures differ with respect to:

1. permeability recovery: a higher recovery is achieved at higher filtration temperatures, with 100% permeability recovery achieved at a 30^{o}C filtration temperature but not at 10^{o}C, and
2. parameters influencing permeability recovery: for NaOCl at a 10^{o}C filtration temperature all parameters influenced recovery, whilst for NaOCl at 30^{o}C only concentration and temperature appeared significant. Moreover, quadratic and interaction coefficients were found to be insignificant for permeability recovery at 30^{o}C.

Table 6-9 Comparison of the percentages of permeability recovery predicted by the model and obtained on the pilot plant

	Parameters applied	Bench test temperature	Pilot plant temperature	%R Model	%R Pilot plant
NaOCl (pH = 10)	[NaOCl] = 600 ppm as Cl Soak temperature = 26^{o}C Soak time = 120 min	10^{o}C	12^{o}C	67.8%	68.5%
NaOCl	[NaOCl] = 600 ppm as Cl Soak temperature = 30^{o}C Soak time = 120 min	10^{o}C	17.5^{o}C	50.6%	63.8%
NaOCl	[NaOCl] = 600 ppm as Cl Soak temperature = 30^{o}C Soak time = 120 min	30^{o}C	21^{o}C	87.1%	83.5%

Comparable observations were made for the pilot plant, in that higher permeability recoveries were obtained at higher filtration temperatures and the influence of the various parameters followed a similar pattern to that observed at bench scale. At filtration temperatures close to those used at bench scale, the pilot model derived from the bench-scale data fitted well with the pilot plant results. The reliability of the bench scale data is thus contingent upon performing the permeability recovery test at the same temperature as that of the feedwater for the pilot plant.

6.4 Conclusions

Box-Behnken design can be used to determine the optimum parameters for the CIP of MF membranes which have been fouled in situ. The impact of different chemical cleaning protocols on permeability recovery has been quantified with respect to reagent type, concentration, temperature and soaking time by providing predictive model equations. The cleaning conditions for each reagent have been optimised to reveal the comparative permeability recovery for each reagent for a single membrane type (PVDF hollow fibre) fouled through operation at constant flux and a specific physical cleaning protocol, and with prefiltration using a specific microscreen.

Results are in agreement with previous studies in that synergistic effects appear important in determining optimum chemical cleaning, and the relative influence of the three key parameters is system dependent. Neither the effect nor the significance of parameters (temperature, concentration and soaking time) follow the same behaviour for each chemical reagent, corroborating previously reported findings. This suggests that the CIP must be optimised for each installation and application, and the the BBD method provides a time-efficient means of doing this.

Sodium hypochlorite adjusted to pH 10 was found to be the most efficient cleaning reagent, yielding a permeability recovery of 100% or more. An

envelope of optimum parameters was defined from the bench scale study for the NaOCl pH 10 and applied to the pilot plant. It was concluded that the bench-scale data could be replicated at the pilot plant scale only if the two sets of data referred to the same temperature.

6.5 References

Bartels, C.R., Franks, R., Furukawa, R., Murkute, P., Papukchiev, U., Integrated membrane system for low fouling RO desalting of municipal wastewater, final report for the Desalination Research and Innovation Partnership, conducted by the City of Oceanside in cooperation with Hydranautics, 2004.

Bartlett, M., Bird, M. R. and Howell, J. A. (1995), An experimental study for the development of a qualitative membrane cleaning model, *Journal of Membrane Science,* 105 (1-2), p. 147-157.

Bohner, H. F. and Bradley, R. L. (1990), Effective Control of Microbial Populations in Polysulfone Ultrafiltration Membrane System, *Journal of Dairy Science,* 73 (9), p. 2309-2317.

Brant, J. A., Kwan, P., Daniel, U. and Valencia, R. (2010), Pilot-scale evaluation of chemical cleaning protocols for organic and biologically fouled microfiltration membranes, *Journal of Environmental Engineering,* 136 (5), p. 542-553.

Chen, J. P., Kim, S. L. and Ting, Y. P. (2003), Optimization of membrane physical and chemical cleaning by a statistically designed approach, *Journal of Membrane Science,* 219 (1-2), p. 27-45.

Heijman, S., Vantighem, M., Raktoe, S., Verberk, J., van Dijk, J. (2007), Blocking of capillaries as fouling mechanism for dead-end ultrafiltration, *Journal of Membrane Science*, 287, p. 119-125

Huang, H., Young, T. and Jacangelo, J. G. (2009), Novel approach for the analysis of bench-scale, low pressure membrane fouling in water treatment, *Journal of Membrane Science,* 334 (1-2), p. 1-8.

Kimura, K., Yamamura, H. and Watanabe, Y. (2006), Irreversible fouling in MF/UF membranes caused by Natural Organic Matters (NOMs) isolated from different origins, *Separation Science and Technology,* 41 (7), p. 1331-1344.

Lazarova, V., Gallego, S., Molina, V.G. and Rougé, P. (2008), *Problems of operation and main reasons for failure of membranes in tertiary treatment systems*, *Water Science and Technology*, 57 (11), p. 1777-1784.

Li, X., Li, J., Fu, X., Wickramasinghe, R. and Chen, J. (2005), Chemical cleaning of PS ultrafilters fouled by the fermentation broth of glutamic acid, *Separation and Purification Technology,* 42 (2), p. 181-187.

Mendoza-Roca, J., Galiana-Aleixandre, M., Lora-Garcia, J., Bes-Pia, A. (2010), Purification of tannery effluents by ultrafiltration in view of permeate reuse, *Separation and Purification Technology*, 70, p. 296-301

Meyers, R.H., Montgomery, D.C., Anderson-Cook, C.M. (2009), *Response surface methodology – Process and product optimization using designed experiments*, 3rd Edition, Whiley, New York.

Mohammadi, T., Madaeni, S. S. and Moghadam, M. K. (2003), Investigation of membrane fouling,, *Desalination,* 153 (1-3), p. 155-160.

Popovic, S., Milanovic, S., Ilicic, M., Djuric, M., Tekic, M. (2009), Flux recovery of tubular ceramic membranes fouled with whey proteins, *Desalination*, 249, p. 293-300

Porcelli, N., Hillis, P. and Judd, S. (2009), Microfiltration membrane plant start up: A case study with autopsy and permeability recovery analysis, *Environmental Technology,* 30 (6), p. 629-639.

Porcelli, N. and Judd, S. (2010a), Chemical cleaning of potable water membranes: A review, *Separation and Purification Technology,* 71 (2), p. 137-143.

Porcelli, N. and Judd, S. (2010b), Impact of cleaning protocol on membrane permeability recovery: a sensitivity analysis, *J. AWWAF*, 102 (12)

Raffin. M., Germain, E., Judd, S. (2011), Optimising operation of an integrated membrane system (IMS) — A Box–Behnken approach, *Desalination*, 273, p. 136-141.

Porcelli, N. and Judd, S. (2010c), Chemical cleaning of potable water membranes: The cost benefit of optimisation, *Water Research*, 44 (5), p. 1389-1398

Strugholtz, S., Sundaramoorthy, K., Panglisch, S., Lerch, A., Brügger, A. and Gimbel, R. (2005), Evaluation of the performance of different chemicals for cleaning capillary membranes, *Desalination,* 179 (1-3), p. 191-202.

Tang X., Flint, S., Benett, S., Brooks, J. (2010), The efficacy of different cleaners and sanatisers in cleaning biofilms on UF membranes used in the dairy industry, *Journal of Membrane Science*, 352, p. 71-75

Tian, J., Chen, Z., Yang, Y., Liang, H., Nan, J. and Li, G. (2010), Consecutive chemical cleaning of fouled PVC membrane using NaOH and ethanol during ultrafiltration of river water, *Water Research,* 44 (1), p. 59-68.

Tran-Ha, M. H. and Wiley, D. E. (1998), The relationship between membrane cleaning efficiency and water quality, *Journal of Membrane Science,* 145 (1), p. 99-110.

Väisänen, P., Bird, M. R. and Nyström, M. (2002), Treatment of UF membranes with simple and formulated cleaning agents, *Food and Bioproducts Processing: Transactions of the Institution of of Chemical Engineers, Part C,* 80 (2), p. 98-108.

Yamamura, H., Kimura, K., Watanabe, Y. (2007), Mechanism involved in the evolution of physically irreversible fouling in microfiltration and ultrafiltration membranes used for drinking water treatment, *Environmental Science and Technology*, 41 (19), p. 6789-6794

Yu, C-H., Fang, L-C., Lateef, S., Wu C-H., Lin, C-F. (2010), Enzymatic treatment for controlling irreversible membranefouling in cross-flow humic acid-fed ultrafiltration, *Journal of Hazardous Materials*, 177, p. 1153-1158.

Zondervan, E. and Roffel, B. (2007), Evaluation of different cleaning agents used for cleaning ultra filtration membranes fouled by surface water, *Journal of Membrane Science,* 304 (1-2), p. 40-49.

CHAPTER 7: ASSESSMENT OF FOULING OF AN RO PROCESS DEDICATED TO INDIRECT POTABLE REUSE

Raffin, M., Germain, M., Judd, S. (2011), Assessment of fouling of an RO process dedicated to indirect potable reuse, Submitted to *Desalination and Water Treatment*.

7.1 Introduction

Increasing freshwater scarcity continues to further the technological progress and economic benefit of wastewater reuse, predominantly to preserve freshwater resources. The use of an integrated membrane system (IMS), the combination of micro/ultrafiltration (MF/UF) followed by reverse osmosis (RO) membranes, represents an important option for municipal wastewater reuse. Such schemes are currently used for advanced treatment of municipal effluents for reuse in industrial processes (Majmaa *et al.*, 2010), environmental protection/restoration (Cazurra, 2008), irrigation (Lazarova *et al.*, 2008) and indirect potable reuse (Markus and Deshmukh, 2010).

A major drawback of such systems is the fouling of RO membranes. Fouling leads to an increase in feed pressure of the system to maintain a constant flow, such that the energy demand also increases. Given that operation beyond some threshold pressure is not tenable, chemical cleaning or dosing for fouling amelioration becomes necessary. Both chemical cleaning and fouling appear to shorten the membrane life, leading to significantly increased operational costs due to membrane replacement (Pointié *et al.*, 2005; Alhadidi *et al.*,2009). It is therefore ultimately necessary to employ appropriate pretreatment to control and/or ameliorate fouling.

Four types of fouling arise on RO membranes: colloidal, biological, organic and inorganic (Bartels *et al.*, 2005). Biofouling of RO membranes can be controlled through ensuring a chloramine residual in the influent (Xu *et al.*, 2010). Organic fouling can be minimised by applying an appropriately conservative flux (Bartels *et al.*, 2005). Inorganic fouling, or scaling, by sparingly soluble salts such as calcium carbonate is suppressed by antiscalant (chemical) dosing, pH reduction and/or reduced recovery (Ghafour, 2002). Colloidal fouling, as well as biofouling, is controlled by pre-treatment (Bartels *et al.*, 2005; Kim *et al.*, 2008). However, notwithstanding pre-treatment, fouling is always experienced to some extent. Autopsy of the RO membrane, whilst providing only a destructive

examination, provides a means of assessing foulants and possible loss of membrane integrity, thereby informing appropriate pre-treatment (Pointié *et al.*, 2005).

This paper provides results from a study of RO membrane autopsies relating to an indirect potable reuse process, along with an appraisal of five antiscalant reagents.

7.2 Materials and Methods

7.2.1 Pilot plant overview

The 600 $m^3.d^{-1}$ Indirect Potable Reuse (IPR) pilot plant has been described elsewhere (Raffin *et al.*, 2011). Final effluent from a conventional activated sludge plant passes through a pre-filter (Bollfilter) before being filtered by a submerged microfiltration (MF) unit (Memcor). The MF permeate then passes through a reverse osmosis (RO) system (Hydranautics) and on to an advanced oxidation process (AOP) and a degasser tower before undergoing pH correction. Chloramine dosing for biofouling control can take place at three different points in the process, pre pre-filter, pre MF and pre RO. Antiscalant and sulphuric acid are dosed pre RO to control scaling. The plant is fully automated and data recorded on a supervisory control and data acquisition (SCADA) system. The RO process has three stages (Table 7-1) and is fed with tertiary MF effluent (Table 7-2).

The RO process is fed from a balance tank by a feed pump at a constant flow rate of 8.2 m^3/h. 20 µm cartidge filters are used to remove any remaining particles to protect the RO membranes. A high pressure pump is used to increase the feed pressure.

Table 7-1 RO process specifications

Manufacturer	Hydranautics
Membrane Type	ESPA2 (1st stage) and ESPA2-4040 (2nd and 3rd stage)
Materials	Composite polyamide
Area/element	37.1 m^2 (ESPA2) and 7.9 m^2 (ESPA2-4040) (leading to an overall area of 365 m^2)
Configuration	Array 1:2:1 (6 elements per vessel) (Total number of elements: 24)
Operating pH	2-10.6
Standard CIP	CIP 1: Recirculation of permeate water at pH 2.5 during 30 min on the 3rd followed by 1hour of soaking CIP 2 and 3: Recirculation of permeate water at pH 2.5 during 30 min on all stages flowed by 1h soaking

Table 7-2 Average RO feed water quality

Parameters	Values
Conductivity (uS.cm-1)	1146 ± 38
TOC (mg/L)	8.0 ± 1.8
pH	7.3 ± 0.1
Temperature (^oC)	13.7 ± 0.88
Turbidity (NTU)	0.04 ± 0.00
Alkalinity (as CaCO3) (mg/L)	195 ± 15
Phosphate (as PO_4^-)	4.09 ± 0.75

7.2.2 Autopsies

Membrane autopsies were conducted on three RO elements: the lead element of the 1st stage, central element of the 2nd stage, and the end element of the 3rd stage, to examine fouling. This was performed after running the process at 85% recovery at a flux of 19 $L.m^{-2}.h^{-1}$ (LMH) and at a pH of 6.5. The antiscalant used was antiscalant A at a dose rate of 2 ppm (Table 7-3). Autopsies comprised optical microscopic investigation, scanning electron microscopy coupled with energy dispersive X-ray (SEM-EDX), chemical analysis by inductively-coupled plasma optical emission spectrometer (ICP-OES), and total cell count determination using DAPI (4,6-diamidino-2-phenylindole) dye staining and fluorescence microscopy. Membrane analyses were performed by IWW

Rheinisch-Westfälishes Istitut für Wasser Beratungs und Entwicklungsgesell-shaft mgH (Germany).

7.2.3 Chemicals

Monochloramine, formed using sodium hypochlorite and ammonium sulphate in a 3:1 mass ratio (N:Cl), was dosed in-line upstream of the pre-filter at a constant dose of 1 mg/L using a static mixer to control biofouling. Antiscalant and acid for scaling inhibition were dosed in-line upstream of the reverse osmosis (RO) process using another static mixer. Previous scoping trials determining optimum operating envelopes for scaling mitigation in the RO process using a single commercial antiscalant (Raffin et al., 2011; Chapter 4) established that both antiscalant and acid dosing were necessary to control scaling. With this antiscalant (Reagent A, Table 7-3), the highest design recovery (85%) demanded adjustment to a pH below 6.25 (by dosing with sulphuric acid at ~ 1.4 l/h) combined with an antiscalant dose of 2 mg/L.

To attempt to reduce sulphuric acid consumption, four different antiscalants (B, C, D and E, Table 7-3), all claimed by the suppliers to be effective against both the phosphate and carbonate salts of calcium, were assessed. Of these, two were commercialised and two were under development. For each antiscalant the appropriate dose, ranging from 2 to 4 mg/L, was determined based on feedwater quality and projections informed by the suppliers' own respective software.

All experiments were conducted under challenging conditions of a mean flux of 19 LMH and an overall recovery of 85%. Experiments were stopped once a 10% decrease in permeate flow for a single stage was observed, since this represents the point at which chemical cleaning is generally advised. Sulphuric acid was used to adjust the pH, which ranged from 6.35 to the natural pH of 7.2.

Table 7-3 Antiscalants properties

Antiscalant	Commercial status	Target scalant	Type
A	Commercialised	Calcium carbonate	Phosphonic acid
B	Commercialised	Calcium carbonate Calcium phosphate	Phosphonic acid
C	Commercialised	Calcium phosphate Calcium carbonate	Phosphonate and carboxylic acid
D	Non-commercialised	Calcium carbonate Calcium phosphate	Unknown
E	Non-commercialised	Calcium carbonate Calcium phosphate	Unknown

7.3 3.Results and discussion

7.3.1 Fouling determination and membrane integrity assessment

Fouling on 1st and 2nd stage elements was observed as a brown, highly hydrated slimy deposit located at the intersections of the spacer material. This deposit was mainly composed of aggregated and suspended bacteria, with a few embedded inorganic particles. For the 3rd stage, a brown-tainted particulate deposit was spread all over the membrane surface and was mainly crystalline inorganic particles, 1 to 40 μm in size. Bacterial aggregates and suspended cells were also observed (Table 7-4) and were more concentrated in Stages 2 and 3 than in Stage 1.

The inorganic component of the fouling layers of the three stages was analysed by ICP-OES (Table 7-4). The deposit analysed at Stages 1 and 2 was found to have a very high water content (~97.5 %) compared with that sampled at Stage 3 (78 %). Data for mass loss on ignition at 550°C revealed the organic content

of the deposits to be 87, 70 and 19 % for Stages 1, 2 and 3 respectively. Carbonates, as detected by loss on ignition at 900 °C, were only present in significant amounts for the Stage 3 deposit, providing 7.5 % of the dry weight.

Table 7-4 Elemental composition of fouling deposits on membrane surfaces determined by ICP-OES and total cell count determined by DAPI staining and fluorescence (< means undetected)

Stage	Stage 1	Stage 2	Stage 3
Wet weight (g/m^2)	3.213	4.901	0.858
Dry weight (g/m^2)	0.077	0.127	0.190
Dry weight (% of wet weight)	2.4	2.6	22.1
Loss on ignition 550^{o}C (% of DW)	86.6	70.1	19.4
Loss on ignition 900^{o}C (% of DW)	87.3	71.9	26.9
Element (ICP analysis) (mg/ m^2)			
Al	0.2256	0.6666	1.1234
Ca	1.4460	9.0671	46.1372
Cd	0.0001	0.0015	0.0063
Co	0.1031	0.0039	0.0266
Cr	0.0257	0.0294	0.0394
Cu	0.0386	0.1397	0.4262
Fe	0.2047	0.2588	0.4725
K	0.1973	<	<
Mg	<	<	0.9605
Mn	0.0039	0.0147	0.0926
Na	0.3856	1.4654	0.5994
Ni	0.0495	0.0093	0.0256
Pb	0.0109	0.9851	0.1852
Total P	1.4107	5.3913	29.1573
Si	0.3406	<	0.9004
Zn	0.0100	0.1245	0.5454
Total cell count/cm^2	3.5×10^6	6.5×10^6	6.7×10^6

The inorganic component of the deposits comprised mainly Ca and P; inorganic phosphates were detected in the deposits from all three RO modules. In addition, Na, Al, Cu, Fe, Mg, Pb, Zn and Si were found at low concentrations. In

comparison, the amounts of Ca and P – most likely from calcium phosphate – were significantly higher on the membrane surface of Stage 3. Stage 2 showed slightly elevated Ca and P concentrations (about 500 times less than for Stage 3) whereas Stage 1 had the lowest Ca and P deposits (2000-3000 times less than Stage 3).

Figure 7-1 compares the inorganic element concentration on Stages 1 and 3, and the expected concentration on Stage 3 if the deposition of inorganic particles is assumed proportional to the bulk retentate concentration, i.e. on a concentration factor of 100/15 between the lead elements of Stage 1 and the tail elements of Stage 3. As shown in Figure 7-1, the concentrations of Ca and P are significantly greater than that based simply on retentate concentration, indicating calcium phosphate ($Ca_3(PO_4)_2$) formation has taken place. According to data in Table 4, a maximum Ca concentration of 40 mg/cm^2 is associated with phosphate. It must therefore be assumed that some calcium carbonate or other calcium-based scales (such as calcium salts of antiscalant) must also be present. It may also be noted that phosphorus can be associated with biomass and the antiscalant itself, especially for Stage 1.

In general, membrane deposits in Stages 1 and 2 were dominated by organic matter (respectively 86.6% and 70.1%) with calcium phosphate making up most of the inorganic component. The Stage 3 deposit was conversely lower in organic content (19.4 %) and higher in concentration of calcium phosphate with some metal carbonates in the inorganic fraction. A similar proportion of scale to organic matter (80%:20%, where most of the scaling was calcium phosphate) was observed by Ning and Troyer (2007), and trends in organic/inorganic content over the three stages were largely in agreement with those reported by Xu *et al.* (2010) from their membrane autopsies of a two-stage pilot-scale reverse osmosis process treating microfiltered municipal wastewater.

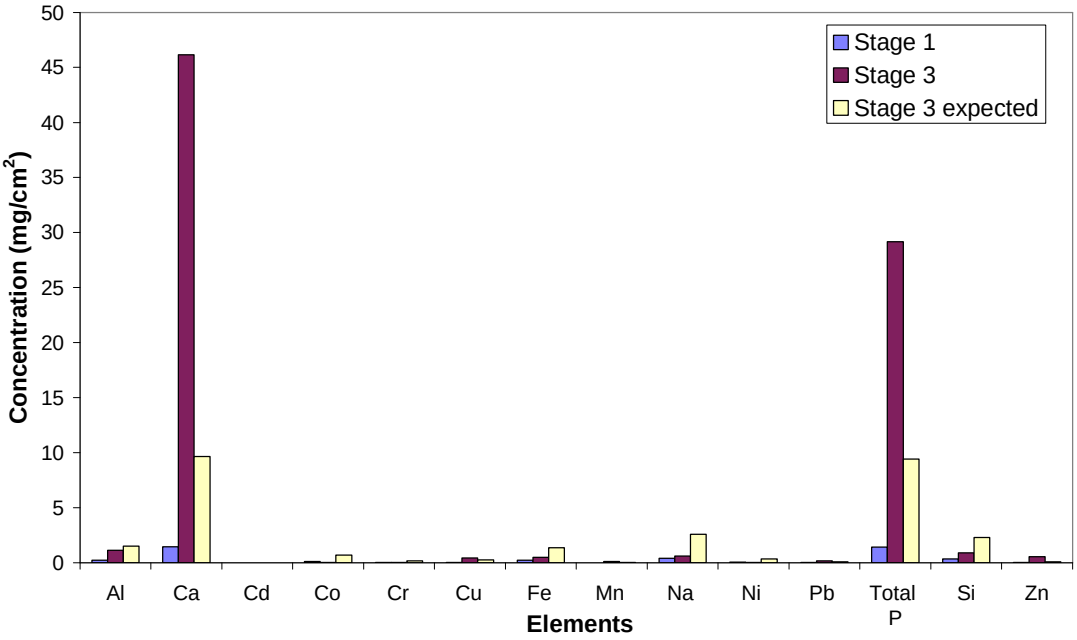


Figure 7-1 Elements concentration (mg/cm^2) for stage 1 and 3, and expected concentration of stage 3 if inorganic fouling follows the concentration factor

Fouling of the RO membrane was, as expected from normal practice, manifested as decreased permeate flow accompanied by decreased salt rejection and increased differential pressure at the third stage of the array. An acid clean at pH 2.5 completely recovered the flow on the third stage. This suggested that, whilst organic and/or biological fouling was evident, scaling was primarily responsible for reduced permeation.

7.3.2 Scaling minimisation

The choice of Antiscalant A was originally made on the assumption of calcium carbonate being the primary scalant, contrary to the outcomes of the autopsy which suggested calcium phosphate scaling to predominate.

Table 7-5 shows that, as expected, the volume of water treated before a chemical cleaning is higher at lower pH. Antiscalant B appeared more efficient

at low pH than Antiscalant A. However at higher pH, Antiscalant B was less efficient. Antiscalant C proved less efficient than Antiscalant A, even though the former is designed for calcium phosphate scaling suppression. It is possible that it was dosed at too high a concentration, causing clogging of the membrane channels. Antiscalants D and E, both of which are under development, provided better results than the commercialised reagents at sulphuric acid acidified pHs. It is likely that at pHs below 6.5 the limiting scaling/fouling potential has shifted from calcium phosphate to some other species yet to be determined.

Table 7-5 Volume of water treated (m^3) before a 10% decrease of the flow on the 3rd stage for each antiscalant as a function of the pH for each tested antiscalant

pH	Volume of water treated (m^3) before a 10% decrease of the flow on the 3rd stage for each antiscalant				
	*A**	*B*	*C*	*D*	*E*
6.35	1247	3865	-	12841	5634
6.5	908	-	843	1114	5556
6.65	430	-	-	-	979
6.75	190	92	-	-	-
7.2	0.104	0.004	0.003	-	-

* Empirical model data based on pilot plant performance data [11]

At the unadjusted pH of 7.2, the RO membranes immediately scaled for all of the commercialised antiscalants. This is contrary to the projection obtained from the antiscalants suppliers' software, which indicated that no pH adjustment was required for pH below 7.6 for the reagents to be effective. Although this was already known for Antiscalant A (Raffin *et al.*, 2011), this insufficiently conservative projected performance was also noted by Xu *et al.* (2010), who reported significant amounts of calcium, aluminium and phosphorus scaling on the membrane whilst projections estimated that only barium sulphate would precipitate without antiscalant. Greenberg *et al.* (2005), who compared five different antiscalants, also reported all tested antiscalants as being ineffective against calcium phosphate scaling when treating secondary wastewater. This may be due to calcium phosphate arising in colloidal form in wastewater effluent

(Ning and Troyer, 2007), such that it passes through the MF and blinds the RO membrane surface; antiscalants are not effective against suspended compounds since they act by suppressing precipitation. Ning and Troyer (2007) also suggested pH control to be critical, since phosphate nanoparticle concentration changes within the pH range of 5-7.

7.3.3 Operating cost

The choice of antiscalant also impacts on the capital and operating costs of the RO process. From the results obtained in the current study, it is evident that the chemical cleaning interval and acid dose required depend on the choice of antiscalant. Cleaning-in-place (CIP) of the RO process can take up to 6 hours depending on the extent of the scaling. At longer CIP intervals the percentage downtime decreases and the net flux increases commensurately, reducing the required number of membrane elements. Acid dosing can be reduced at lower recoveries. There is therefore a trade-off between various design and operating parameters and, according to the results, it appears that the operating envelope defined by Raffin *et al*. (2011) could be enlarged. The reduction of acid dosing also impacts favourably on site health and safety issues relating to sulphuric acid storage.

A sensitivity analysis was performed to assess the operating cost saving arising from chemical dosing (pH and antiscalants dose). Ranges and costs of the parameters used for the sensitivity analysis are summarised in Table 7-6. Figure 7-2 represents the contribution of acid and antiscalant to chemical costs as a function of the adjusted pH. Similar trends are obtained with the other antiscalants. Costs of chemical dosing range from £0.008/m^3 feed water for zero acid dosing and 2 mg/L of antiscalant to £0.040/m^3 feed water when dosing to a pH of 6.25 and antiscalant dose of 4 mg/L. On average, the operating cost involves by chemical dosing is decreased by 7.8% for each 0.1 unit increase in the adjusted pH, which correspond to a decrease of £0.003/m^3. An additional 0.55% (~0.0002 £/m^3) reduction arises with each 0.1 mg/L

decrease in antiscalant dose. However, the cost contribution from the acid and the antiscalant depends on their respective doses: the higher the dose of antiscalant the higher the pH that can be sustained and the greater the contribution of antiscalant cost to the overall operating cost (Figure 7-2). Over the range of conditions studied, pH adjustment was found to have the greatest influence on operating cost, with possible operational cost reductions of 67-77% for zero acid dosing compared to adjustment to pH 6.25.

Table 7-6 Ranges and prices of the different parameters

	Parameters Range	Reagent price (£/kg)
Sulphuric acid (mg/L)	0 – 185*	0.17
Antiscalant (mg/L)	2 – 4	1.4 – 4

*Corresponding pH range: 6.25 – 7.2.

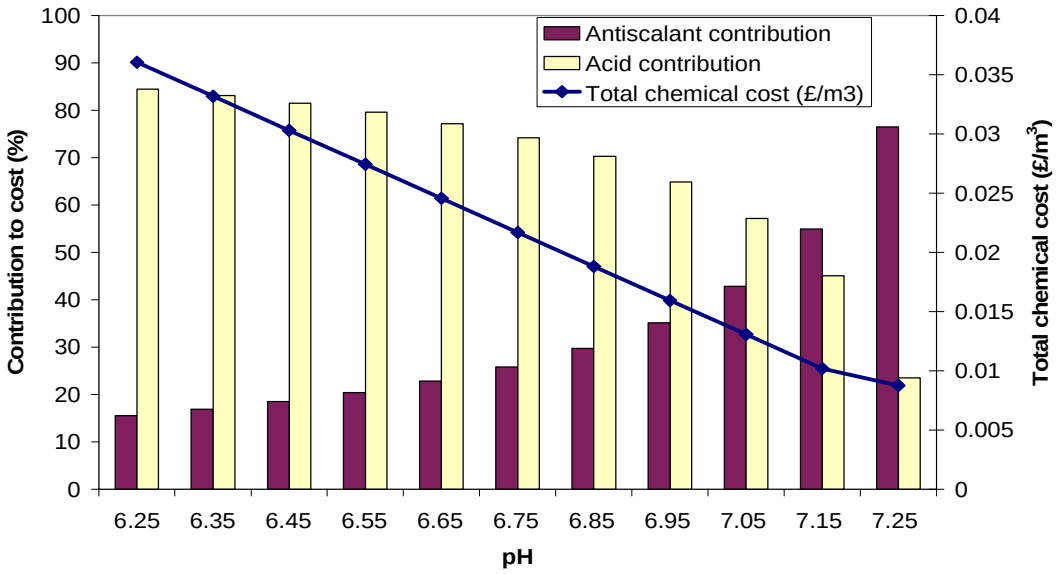


Figure 7-2: Contribution of acid and antiscalant to cost and total chemical cost as a function of the adjusted pH (from a pH of 7.25 and an alkalinity of 195 mg/L as $CaCO_3$) for Antiscalant *A* (concentration of 2 mg/L)

7.3.4 Biofouling minimisation

In the existing scheme biofouling is minimised by pre-treating the RO influent by MF, along with dosing to 1 mg/L with chloramine. However, this pretreatment was found not to eliminate microbial activity in the RO feedwater, where a colony count at 22 ^{o}C of ~180/ml was recorded. As reported by Lazarova *et al.* (2008), low molecular weight dissolved organics passed through the microfiltration membrane and provide nutrients for micro-organisms immobilised in biofilms. In this study, the DOC concentration reached up to 10 mg/L in the RO feed water. Villacorte *et al.* (2009) showed that a biofilm may result from the deposition of transparent exopolymer particles arising from pre-treatment. The concentration of phosphate in the RO feed water, especially when treating wastewater, may also contribute to biofouling. Vrouwenvelder *et al.* (2010) demonstrated that reduced phosphate concentrations can significantly constrain biomass accumulation, and it is well known that phosphonate-based antiscalants may promote RO biofouling by increasing phosphate concentration in the presence of an organic carbon substrate. These authors advised limiting phosphate levels by implementing pre-treatment such as coagulation, and avoiding phosphonate-based antiscalants when treating wastewater effluents rich in organic substrate. Organic polymer-based antiscalants with highly assimilable organic compounds were also found to have a high biofouling potential by providing nutrients to micro-organisms (2000).

Volatile organic foulant was found on membranes in all stages, portions of which are likely to derive biological growth. A higher bacteria count per square centimetre was recorded for the tail elements of the 3rd stage (Table 7-4), corroborating the reported results of Xu *et al.* (2010). According to these authors, the chloramine residual decreased along the length of the module and between successive stages since its rejection by the RO membrane is low. In the current study the chloramine concentration in the RO permeate was found to be higher than that recorded in the feed water, with no residual chloramines in the retentate. Clearly, the impact of chloramines dosing on biofouling

mitigation in the concentrate scheme would be expected to be negligible under such conditions.

7.4 Conclusion

Membrane autopsies have been conducted to assess fouling propensity of a RO membrane process treating wastewater effluent, along with pilot trials of a range of antiscalants which were compared with reference to scaling mitigation.

Autopsies showed the first and the second stages of an RO plant treating microfiltered secondary municipal wastewater to be subject to significantly less scaling than the third stage, respectively 15 and 4 times less than Stage 3. Scaling was mostly associated with calcium phosphate, although calcium carbonate was also present. Biofouling was observed on all three stages with higher concentrations at the 3rd stage. This was explained by a lack of chloramine residual since its concentration decreases across the array.

Results show that antiscalant efficiency regarding scaling minimisation differs between products. A future generation of antiscalants are being developed and showed some promising results. However, no commercial antiscalant appears capable of avoiding scaling without the addition of sulphuric acid. This might reflect by the relative inefficacy of these reagents against calcium phosphate colloidal fouling.

A simple analysis enabled different antiscalants to be appraised. A cost analysis quantified the benefit of employing a more effective antiscalant at more neutral pH levels: a small increase in adjusted pH can significantly reduce operational costs associated with acid consumption. An operating cost reduction of up to 77% can be obtained by increasing adjusted pH from 6.25 to 7.25 at wastewater alkalinity of 195 mg/L as $CaCO_3$. Capital costs might also be reduced since slightly fewer membrane elements are required at longer chemical cleaning intervals and commensurately higher net fluxes.

From this study, it is clear that more attention is required regarding pre-treatment of the RO process to limit different types of fouling.

7.5 References

Alhadidi, A., Kennedy, M., Diepeven, A., Prummel, H., Boorsma, M., Schippers, J.C. (2009), Scaling potential calculations using different methods, *Desalination and Water Treatment*, 6, p. 138-143.

Bartels, C.R., Wilf, M., Andes, K., Iong, J. (2005), Design considerations for wastewater treatment by reverse osmosis, Water Science and Technology, 51 (6–7), p. 473–482.

Cazurra, T. (2008), Water reuse of south Barcelona's wastewater reclamation plant, *Desalination*, 218, p. 43-51.

Ghafour, E.E.A. (2002), Enhancing RO system performance utilizing antiscalant, *Desalination*, 153, p. 149–153.

Greenberg, G., Hasson, D., Semiat, R. (2005), Limits of RO recovery imposed by calcium phosphate precipitation, *Desalination*, 183, p. 273-288.

Kim, J., DiGiano, F.A., Reardon, R.D. (2008), Autopsy of high-pressure membranes to compare effectiveness of MF and UF pre-treatment in water reclamation, *Water Research*, 42, p. 697-706.

Lazarova, V., Gallego, S., Molina, V.G., Rougé, P. (2008), Problems of operation and main reasons for failure of membranes in tertiary treatment systems, *Water Science and Technology*, 11, p. 1777-1784.

Majamaa, K., Aerts, P.E.M., Groot, C., Paping, L.L.M.J., Van den Broek, W., Van Agtmaal, S. (2010), Industrial water reuse with integrated membrane system increases the sustainability of the chemical manufacturing, *Desalination and Water Treatement*, 18, p. 17-23.

Markus, M.R., Deshmukh, S.S., An innovative approach to water supply – the groundwater replenishment system, In: *World Environmental and Water Ressources Congress 2010: Challenges of Change*, Providence, Rhode Island 16-20 May 2010, p. 3624-3639.

Ning, R.Y., Troyer, T.L. (2007), Colloidal fouling of RO membranes following MF/UF in the reclamation of municipal wastewater, *Desalination*, 208, p. 232-237

Pointié, M., Rapenne, S., Thekkedath, A., Duchesne, J., Jacquemet, V., Leparc, J., Suty, H. (2005), Tools for membrane autopsies and antifouling strategies in seewater feeds: a review, *Desalination*, 181, p. 75-90.

Raffin, M., Germain, E., Judd, S. (2011), Optimising operation of an integrated membrane system (IMS) — A Box–Behnken approach, *Desalination*, 273, p. 136-141.

Villacorte, L.O., Kennedy, M.D., Amy, G.L., Schippers, J.C. (2009), The fate of transparent exopolymer particles (TEP) in integrated membrane systems: Removal through pre-treatment processes and deposition on reverse osmosis membranes, Water Research, 43, p. 5039-5052.

Vrouwenvelder, J.S., Beyer, F., Dahmani, K., Hasan, N., Galjaard, G., Kruithof, J.C., Van Loosdrecht, M.C.M. (2010), Phosphate limitation to control biofouling, *Water Research*, 44, p. 3454-3466.

Vrouwenvelder, J.S., Manolaraskis, S.A., Veenendaal, H.R., Van der Kooij, D. (2000), Biofouling potential of chemicals used for scale control in RO and NF membranes, *Desalination*, p. 132, 1-10.

Xu, P., Bellona, C., Drewes, L.E. (2010), Fouling of nanofiltration and reverse osmosis membranes during municipal wastewater reclamation: Membrane autopsy results from pilot-scale investigations, *Journal of Membrane Science*, 353, p. 111-121.

CHAPTER 8: LIFE CYCLE COST ANALYSIS (LCCA) FOR AN INDIRECT POTABLE REUSE SCHEME – FROM PILOT PLANT TO POTENTIAL FULL-SCALE PLANT

Raffin, M., Germain, E., Judd, S., Life Cycle Cost Analysis (LCCA) for an Indirect Potable Reuse Scheme – From pilot plant to potential full-scale plant, In preparation for submission to *Desalination*.

8.1 Introduction

Increasing freshwater scarcity is making reclamation of wastewater effluent more economically attractive as a means of preserving freshwater resources. The use of an integrated membrane system (IMS), the combination of micro/ultra-filtration (MF/UF) followed by reverse osmosis (RO) membranes, represents a key process for municipal wastewater reuse; it is currently used for advanced treatment of municipal effluents for reuse in industrial processes (Majamaa et al., 2010), environmental protection/restoration (Cazurra et al, 2008), irrigation (Lazarova et al., 2008) and indirect potable reuse (Markus and Deshmukh, 2010).

Membrane processes offer the advantage of improved water quality, and reduced footprint, chemical demand and waste generation over traditional physical/chemical treatment process (Juang et al., 2006); MF has been demonstrated to offer an economically viable alternative to conventional lime clarification/filtration pre-treatment for reverse osmosis (Won and Shields, 1999). However, a survey of the state of the art in IPR technology reveals widely varying operational costs, with no apparent consistent basis for this (Chapter 2). While plant capacity may impact on operational cost to some extent, operating parameters such flux for the MF/UF and flux, recovery and total dissolved solids may significantly impact on operating cost. Only a more rigorous life cycle cost analysis (LCCA) is able to quantify costs and their sensitivity to plant operation and maintenance (O&M) parameters.

A life cycle cost analysis (LCCA) is designed to assess the overall cost of the project from “cradle-to-grave”, are commonly used to select the most sustainable process design and operation (Won and Shields, 1999). In this study, LCCA was applied to two process treatment schemes to assess their relative cost. Both schemes aimed to generate high quality water by applying MF to “secondary” wastewater from a municipal wastewater treatment works, i.e. wastewater discharged from the biological treatment stage. Post treatment

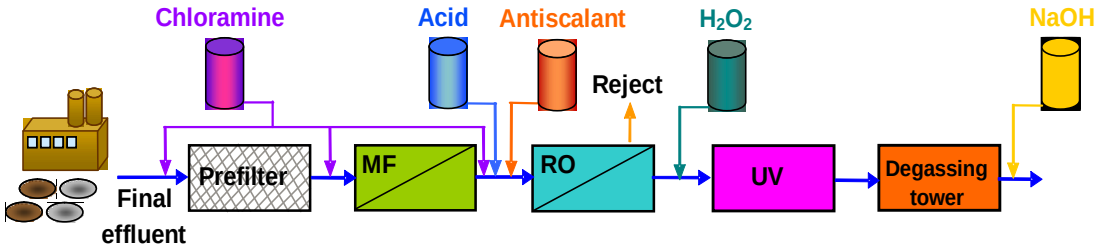
cases included an advanced oxidation process (AOP) based on ultraviolet radiation with hydrogen peroxide dosing (UV/H_2O_2). In the first scheme the AOP was considered to directly follow the MF (MF/AOP), whereas in the second it was preceded by a reverse osmosis (RO) step. All data for plant operating parameters were obtained from operation of a pilot plant, and capital cost data from commercial technology suppliers.

8.2 Material and methods

8.2.1 Treatment trains

The two process treatment trains, *T1* (MF/RO/AOP) and *T2* (MF/AOP), considered in this study (Figure 8-1) were both designed to remove ostensibly potentially onerous organic matter. LCCA was conducted at three different scales of plant capacity: 600 $m^3.d^{-1}$ (i.e. pilot plant), and 25,000 and 100,000 $m^3.d^{-1}$ (full-scale). The design was based on that of a previously reported pilot plant described elsewhere (Chapter 3, Raffin et al., 2011), and in more detail in Appendix 3. The plant operating parameters adopted for the analyses Table 8-1) were based on those defined through optimisation of the 600 $m^3.d^{-1}$ pilot plant with reference to either minimal fouling and/or maximum contaminant removal (Raffin et al., 2011a, 2011b; Hatt et al., 2011 ; James et al., 2011).

Train 1 (T1)



Train 2 (T2)

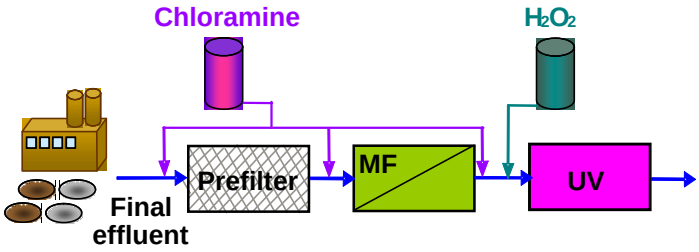


Figure 8-1 Treatment trains, T1 (MF/RO/AOP) and T2 (MF/AOP)

Table 8-1 Process description and operating parameters

Process	*Process description*	*Operating parameters*	*References*
Pre-filter	Automatic Backflush pre-filter	Mesh size: 500 µm Backwash flow: 8.5 $m^3.h^{-1}$ for 20s	Hatt et al. [11]
Chloramine		Chloramine dose 1 ppm	
Microfiltration (MF)	Submerged PVDF hollow fibre membrane	Flux: 56 LMH* Backwash interval: 15 min Backwash: Air (0.40 m/h for 55 s) + water (0.06 m/h for 15 s). Backwash downtime of 300 s NaOCl CIP: 600 ppm as Cl, 30 min recirculation, 90 min soak at 35°C	Raffin et al. [10] Raffin et al. [12]
Reverse osmosis (RO)	3-stages RO process	Flux: 19 LMH* Recovery: 75% pH: 6.75 Antiscalant dose: 2 ppm CIP 1: Recirculation of permeate water at pH 2.5 for 30 min on the 3rd stage followed by 1 h soak CIP 2 and 3: Recirculation of permeate water at pH 2.5 for 30 min for all stages followed by 1 h soak	Raffin et al. [10]
Advanced oxidation process (AOP)	UV + H_2O_2 Low pressure reactor	<u>After MF</u> Power: 100% Hydrogen peroxide dose: 16 mg/L <u>After RO</u>: Power : 60% Hydrogen peroxide dose: 3 mg/L Design and other operating parameters provided by manufacturer	James et al. [13]
Post-treatement	Degassing tower Sodium hydroxide	pH after NaOH dosing: 7-8	

*LMH : $L.m^{-2}.h^{-1}$

8.2.2 Water quality

The mean water quality as recorded in the course of the pilot plant studies is reported in Table 8-2: the same water quality was assumed for the analyses conducted on the full-scale plants. The feed water quality recorded for the pilot plant investigation was found to be reasonably consistent throughout the year,

albeit with some variation in temperature whose impact (ostensibly on water viscosity) was not considered to depend on plant capacity.

Performance of treatment train in terms of compounds removal was not taken into account in this study. However, effluent water quality from T1 is higher than that provided from T2 with respect microbiological, organic and inorganic compounds due to the additional RO membrane step.

Table 8-2: Mean measured feed water quality (2008-2010)

Parameter	*Average*	*Min*	*Max*
Turbidity (NTU)	6.18 ±3.35	0.37	100
TOC ($mg.L^{-1}$)	7.18±0.82	5.82	8.88
Temperature (^oC)	16.7 ± 1.97	8.56	26.54
pH	7.09 ± 0.35	6.55	7.85
Conductivity ($\mu S.cm^{-1}$)	1048 ± 90	630	1862
Alkalinity ($mg.L^{-1}$ as $CaCO_3$)	196 ± 14.6	141	235
UV_{254}	0.196±0.018	0.175	0.256
Specific UV absorbance ($m^{-1}.mg^{-1}.L$)	2.82±0.45	2.14	4.35

1.1 Life cycle cost analysis (LCCA)

The LCCA followed the British Standard (BS ISO 15686-5:2008). The scope of the analysis of the two trains *T1* and *T2* (Figure 8-1) encompassed the construction cost (primarily equipment capital expenditure (CAPEX), installation and commissioning), the operational and maintenance costs (operational expenditure (OPEX) from power, materials and consumables, monitoring/software, labour, maintenance and service contracts). Decommissioning costs were excluded. All costs were obtained from the equipment suppliers, with mean values taken when more than one datum was provided by different suppliers.

The useful life assumed for the plant and the principal components was 30 years for the plant itself, 10 years for the pumps, 7 years for the microfiltration membranes and 5 years for the reverse osmosis membrane. These values were considered to be independent of plant capacity.

Concentrate streams from both membrane processes were assumed to be treated either at the same wastewater treatment plant (WWTP) as the installed plant (in the case of the 600 m^3/d installation) or discharged to sewer to be treated by some other works for the larger installations. This assumption was made since recycling of the majority of the flow to the heads of the same works would lead to unacceptable accumulation of recalcitrant species. It was assumed that the distance between the works and the discharge point to another works was six kilometres.

Labour for each plant was assumed to consist of three manager(s)/employee(s) for 600 m^3/d plant, four for the 25 MLD plant and 9 for the 100 MLD. The unit treatment processes (membrane and advanced oxidation) and instrumentation were assumed to be serviced twice a year.

A number of aspects were ignored in order to provide a consistent analysis. The recovered water was assumed to retain no value other than the environmental benefit, which pertains to the increase in freshwater resource. Thus no discount rate was applied for the recovered water. Whilst the pumping of the wastewater to the works was taken into account, the cost of wastewater treatment (i.e. at conventional primary and secondary treatment) was ignored. Inflation was also ignored, since its impact could reasonable be assumed to be roughly the same across all scales of operation.

8.2.3 Sensitivity analysis

The sensitivity of the calculated costs for LCCA of *T1* to membrane operating parameters (flux and backwash frequency for the MF, and flux and recovery for the RO) was assessed. Values of parameters used for the sensitivity analysis are reported in Table 8-3 and correspond the extreme and normal operating parameters defined by Equations 4-6 and 4-7 in Chapter 4 (Raffin et al., 2011a). These equations represent the limit between fouling and non-fouling, as indicated by the determined operating envelope.

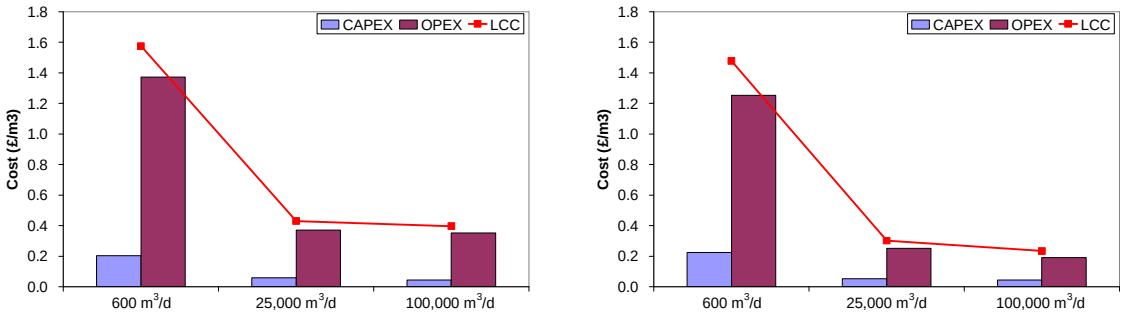
Table 8-3: Sensitivity analysis parameters values

Process	*Operating parameters*		
MF	Flux (LMH)	BW interval (min)	
	56	15	
	45	30	
	32	45	
RO	Flux (LMH)	Recovery (%)	pH
	19	75	6.75
	19	80	6.5
	19	85	6.25
	16	75	6.75
	16	80	6.5
	16	85	6.25

8.3 Results and discussion

8.3.1 Life cycle cost analysis (LCCA)

Life cycle cost (LCC) was calculated for both streams *T1* and *T2* at the three different plant capacities (Figure 8-2). An LCC ranging from £0.40 to £1.57 per m^3 treated water was calculated for *T1* compared with £0.23-1.48/m^3 for *T2*, the cost decreasing with increasing plant capacity. This is an intuitive trend, reflecting the economy provided by operation at larger scale, and widely reported in many studies including those of membrane systems (Coté et al., 2004; Wilf et al., 2010). Data from the current study were of a similar magnitude to those published previously for wastewater reuse using RO (0) for the two full-scale plants. However, the calculated LCC for *T1* was consistently higher than that reported in previous studies (Coté et al., 2004; Wilf et al., 2010).



(a) (b)

Figure 8-2 LCC, OPEX and CAPEX as a function the plant capacity, (a) T1 (MF/RO/AOP) and (b) T2 (MF/AOP) for the 3 plants

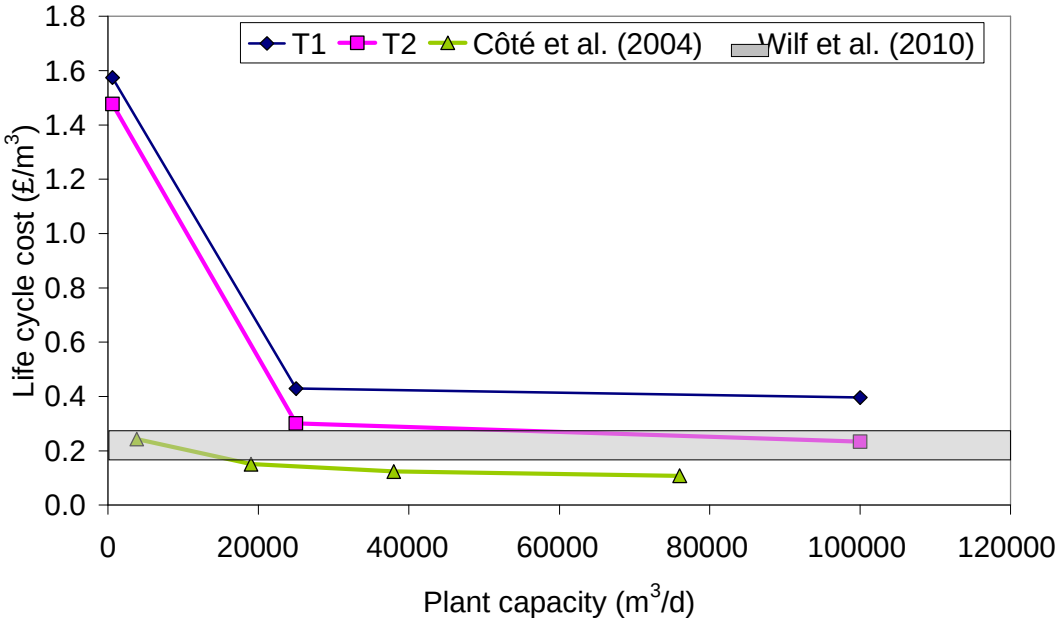


Figure 8-3 Comparison of *T1* and *T2* LCC with literature data.

The CAPEX calculated for the current study for both trains varied from £0.04 (*T2*, 100,000 $m^3.d^{-1}$) to £0.22/m^3 (*T1*, 600 $m^3.d^{-1}$). CAPEX data provided by Côté et al. (2004) varied between 0.05 to 0.10 £/m^3 for plant capacities of

76,000 to 3,800 $m^3.d^{-1}$, comparable with the CAPEX found for both trains for the 25,000 and 100,000 $m^3.d^{-1}$ plants. This indicates that differences in LCC across the different plant capacities and across studies (Coté et al., 2004; Wilf et al., 2010) arise largely from the OPEX. Such disparities arise from differences in assumptions between studies, as well those relating to the treatment train. Whilst previous studies took account of the pre-treatment to the MF/RO process, post-treatment (such as AOP and pH correction in the current study) was excluded, and concentrate disposal costs were also ignored (Coté et al., 2004). Finally, the assumed electrical power costs was considerably lower for the two cited studies - around £0.054/kWh (Coté et al. 2004) compared to £0.11/kWh assumed for the current study. The higher CAPEX for the 600 $m^3.d^{-1}$ plant is attributable to the acquisition of items offering economy of scale. The process and the instrumentation CAPEX values are respectively 3 times and 42 times higher for the smaller plant than those for the 25 MLD, and 3.5 times and 84 times higher than those for the 100 MLD. For larger scale, tanks, membrane vessels and UV reactors are larger, demanding less construction materials per unit volume of treated water. Furthermore, instrumentation per treatment train is unaffected by the flow treated.

Results show that CAPEX and OPEX respectively represent 11 ± 2.5 % and 89 ± 2.5 % of the LCC for *T1* and compared to 15 ± 4 % and 85 ± 4 % for *T2*. Such trends differ somewhat from those reported by Côté et al. (2004) who reported a 25% / 75 % distribution between the CAPEX and OPEX, and of Wilf 2010), who reported a 45% / 55% distribution. Both CAPEX and OPEX decrease with plant capacity reflecting the economy of scale (Sections 8.3.2-3).

The LCC was found to be consistently lower for *T2* by 6.5, 30 and 41 % respectively for the 600 $m^3.d^{-1}$, 25,000 $m^3.d^{-1}$ and 100,000 $m^3.d^{-1}$ plants, although the absolute difference between the *T1* and the *T2* LCC were quite consistent (respectively £0.10, £0.13 and £0.16 at the three different scales). The lower LCC for *T2* is mainly due to the OPEX difference between *T1* and *T2*, and in particular the decrease in energy, chemical and spare parts cost linked with the RO process. Furthermore, since the overall conversion of train *T2* is

higher is the absence of the RO process the energy consumption required for the raw water pumping is lower since the demand of influent is decreased.

8.3.2 Capital costs

Capital cost can be categorised according the contribution from the process (pumping stations, membrane processes, chemical dosing, advanced oxidation process and post-treatment), monitoring, building, electrical work, pipework & fittings, and others (site preparation, health and safety, potable water connection, training and insurance). The contribution from each component was calculated at each plant capacity and for each train (Figure 8-4). For both *T1* and *T2*, process acquisition was found to contribute most significantly to CAPEX (respectively 66 %, 40 % and 59 % on average for the 600, 25,000 and 100,000 $m^3.d^{-1}$ plants) followed by civil work, including building construction and pipework & fittings.

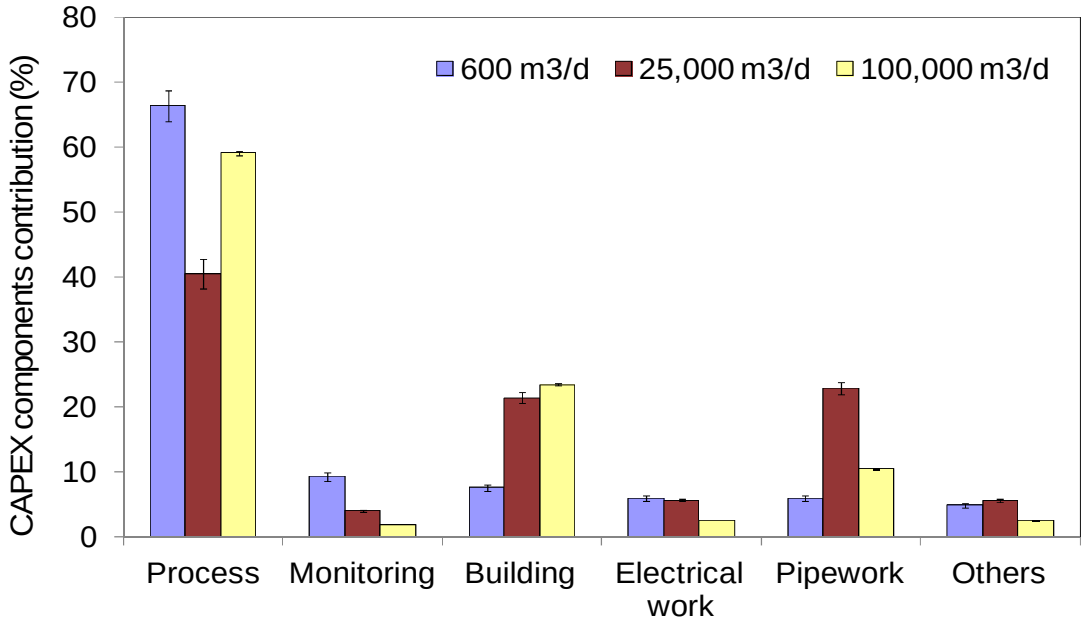


Figure 8-4 Average CAPEX component contribution for T1 (MF/RO/AOP) and T2 (MF/AOP) for the 3 plants

The greater contribution from the pipework & fittings to the cost of the 25,000 $m^3.d^{-1}$ plant compared to the larger and smaller plant arises from the requirement to construct a new sewer to manage the RO concentrate. For the pilot plant this is not required, provided the wastewater treatment works providing the feedwater could be assumed to have a much larger capacity than the pilot plant: the concentrate can simply be returned to the works without adversely impacting on the operation of the latter. For the two larger plants this is not possible, and dedicated concentrate management is required, significantly adding to cost.

Conversely, the proportional cost of monitoring and electrical work decreases from just below 6% of the total cost in the case of the 600 $m^3.d^{-1}$ plant to less than 0.5% for the largest plant. This is again intuitive, since this requirements remain the same and increase only marginally in absolute terms with increasing plant size. Although the proportion of the cost provided by the building housing the plant seems to increase at the largest scale considered (22%, compared to 8% for the pilot plant), the cost per cubic metre is almost unchanged over the capacities studied (£ 0.021±0.001/m^3) and reflects the decreased costs of instrumentation and processes with plant capacity, yielding a higher proportional contribution from the building.

The contribution of the cost of each treatment process to the overall CAPEX was assessed for the two streams at the different plant capacities (Figure 8-5). As expected, the MF and RO membrane processes provide the largest contribution to the *T1* treatment scheme CAPEX, respectively 34 and 40% in average, with the AOP contributing only to 8%. However, for *T2* the AOP provide a significantly greater contribution to the process CAPEX than the MF process, 53% compared with 34%. This arises from the increased number of UV reactors required for *T2* to achieve the same performance as that from *T1*. The reduced transmissivity (~68% according to James et al., 2011) of the permeate from the MF compared with that of the RO (~100%) demands an eight-fold increase in the number of UV reactors required based on the ability to

remove the most recalcitrant of dissolved organic materials down to the levels demanded to achieve the water quality standard (Mackey *et al.*, 2001).

The contribution of the pumping station cost, including pumping of the final effluent, reclaimed water and membrane concentrate pumping station, was found to be higher for the 25,000 and 100,000 $m^3.d^{-1}$ plant. This arises from the significant contribution of membrane concentrate pumping capital equipment for the larger plant.

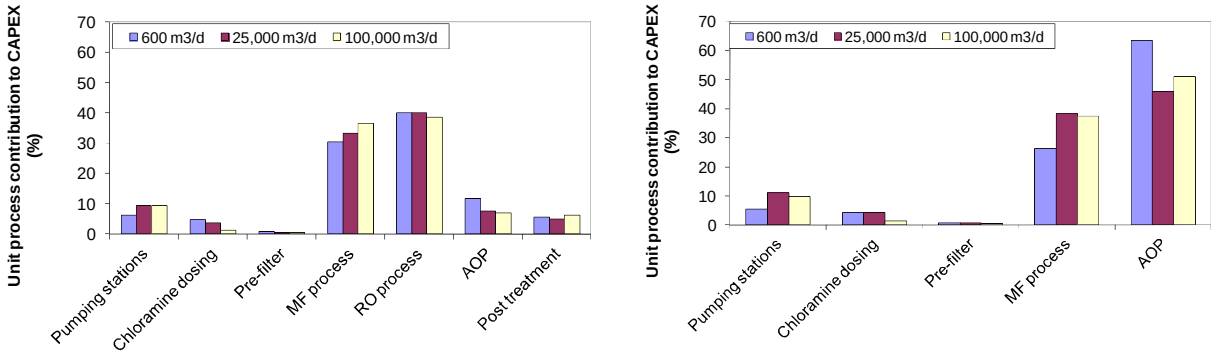


Figure 8-5 Unit process contribution to CAPEX, (a) T1 (MF/RO/AOP) and (b) T2 (MF/AOP)

8.3.3 Operational costs

Operational cost was categorised according to the contribution of processes (pumping stations, membrane processes, chemical dosing, advanced oxidation process and post-treatment), monitoring, building, labour and maintenance and service contracts. The proportional contribution from each component cost was calculated at each plant capacity and for each process treatment train (Figure 8-6). Two key components appear to dominate regarding the contribution to OPEX: labour costs for the 600 $m^3.d^{-1}$ installation, representing 76% of the OPEX, and process operating cost for the 25,000 and the 100,000 $m^3.d^{-1}$ plants, which provide ~70% for the 25,000 $m^3.d^{-1}$ plant and over 90% of the 100,000 $m^3.d^{-1}$ operating costs. This again reflects the significant economy of scale of

the larger plants with respect to the staffing level, the number of staff and servicing decreasing with scale.

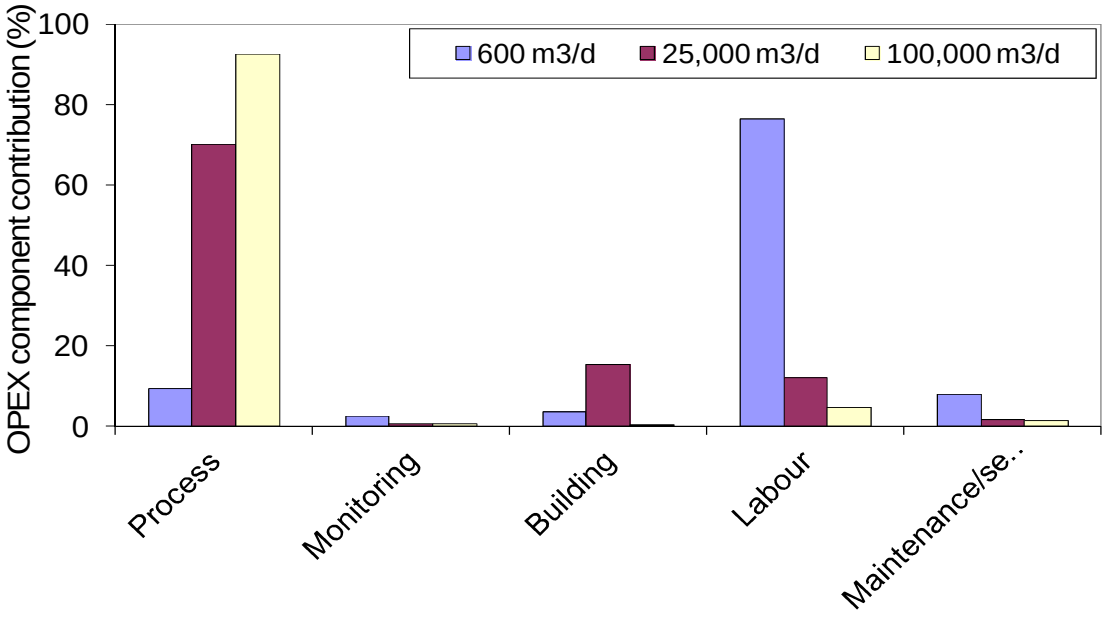


Figure 8-6 OPEX component contribution, (a) T1 (MF/RO/AOP) and (b) T2 (MF/AOP)

As for the CAPEX, the contribution to the process OPEX has been assessed by process component contribution (Figure 8-7). For *T1* the RO provides the highest contribution to the OPEX (42%) followed by the MF (22%). This arises from the greater demand for energy, chemicals and membrane replacement for the RO compared to all the other treatment processes: the RO process contributes 35% of the total energy consumption, 80% the chemical consumption and 60% of the spare parts replacement. For *T2*, the MF provides the highest contribution (42%) to the process OPEX for the two large plants, followed by the AOP which contributes 30% of the running cost. For the 600 $m^3.d^{-1}$ plant the AOP and MF contributions to OPEX were similar at 34-35%, since UV lamp replacement costs per m^3 treated water are higher for a small reactor as employed at 600 $m^3.d^{-1}$ flow capacity. The proportional wastewater

pumping contribution was found to be higher at larger scale due to the requirement of transporting the concentrate to the wastewater treatment works.

Energy consumption provided the highest contribution to the OPEX for *T1* at all scales: 58% for the pilot plant increasing to 83% and 87% for the 25,000 and 100,000 $m^3.d^{-1}$ plants respectively. The predominance of the cost of energy over other contributors to OPEX is expected and has been widely reported for membrane technologies generally and RO processes in particular (Coté *et al*., 2004; Wilf, 2010; Pearce, 2008). Energy consumption increases from 0.8 $kWh.m^{-3}$ for the 600 $m^3.d^{-1}$ to 2.4 $kWh.m^{-3}$ for the 100,000 $m^3.d^{-1}$ plant. These figures are somewhat higher than those proposed by Pearce (2008) for integrated membrane system for wastewater reuse, which ranged from 0.5 to 0.7 kWh/m^3 and included pre-treatment but excluded post treatment which, according to the current study, contributes 0.1-0.2 kWh/m^3. However, the energy consumption found in this study are comparable to those of existing full-scale plants (Chapter 1).

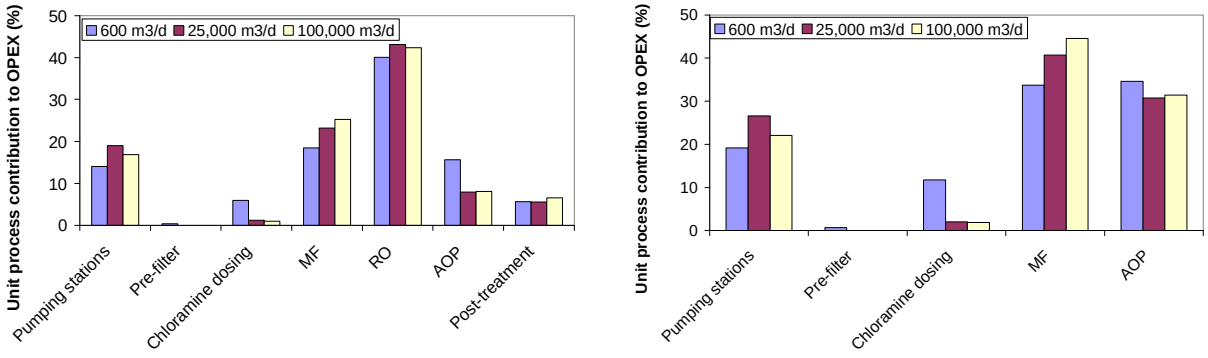


Figure 8-7 Unit process contribution to OPEX, (a) T1 (MF/RO/AOP) and (b) T2 (MF/AOP)

8.3.4 Sensitivity analysis

Sensitivity analysis was performed to assess the effect of operating parameters on the CAPEX and OPEX (Figure 8-8 and Figure 8-9). From Figure 8-8, it can

be concluded that the RO flux exerts the greatest influence on the CAPEX, since decreasing the design flux commensurately increases the membrane area demand. CAPEX also increases with decreasing MF flux and commensurately increasing membrane area. Finally, and as expected, the CAPEX decreases with an increase of the RO recovery as the number of MF and the RO modules required is reduced. From Figure 8-9, the RO recovery has the largest impact on OPEX, with OPEX increasing with the increasing recovery and the concomitant increase in energy demand associated with the higher feed pressure required. A decrease in RO flux increases OPEX since the cost of membrane replacement is increased with the number RO modules. However, this is questionable since, in practice, membrane life decreases with increasing flux.

In the case of the MF, it appears that operation at moderate flux and backwash frequency provides lower OPEX. The air compressor energy consumption is quite high when the BW interval is low (15 min). At low flux (32 LMH), the number of modules is higher than for higher flux and leads to increased OPEX from required MF membrane replacement. However, as with RO, the MF membrane replacement is likely to be related to flux.

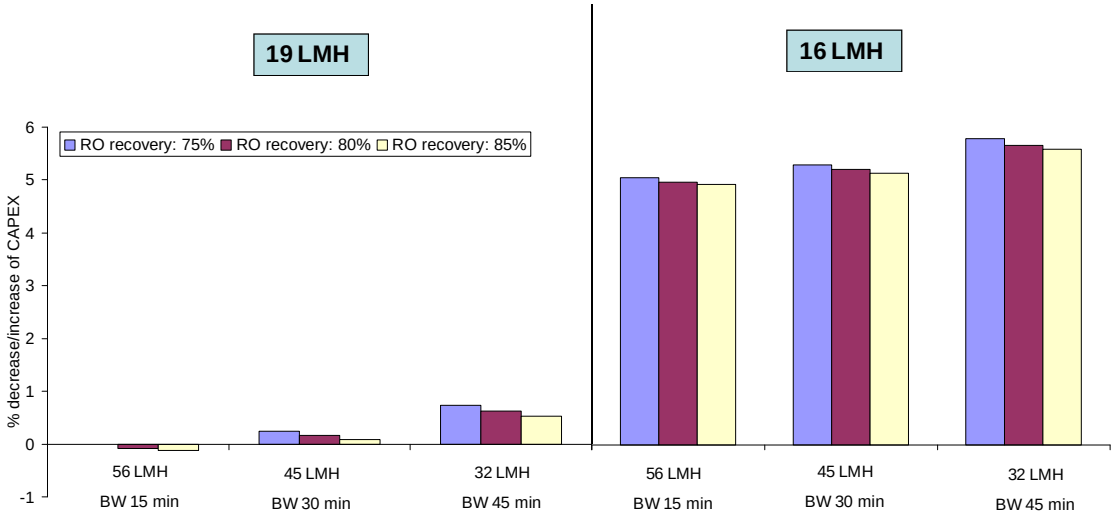


Figure 8-8 Sensitivity of CAPEX to recovery (MF and RO) and RO flux

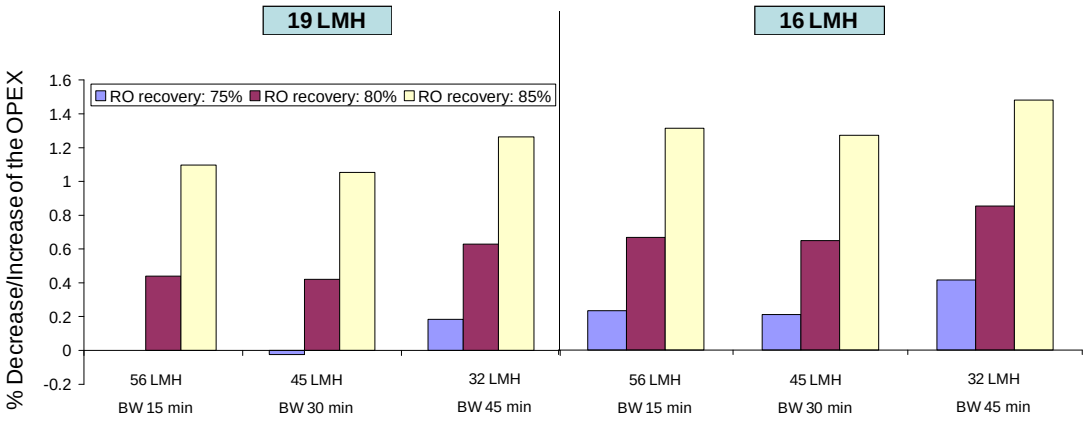


Figure 8-9 Sensitivity of OPEX to recovery (MF and RO) and RO flux

8.4 Conclusions

A life cycle cost (LCC) analysis was performed to compare two trains, *T1* (MF/RO/AOP) and *T2* (MF/AOP) on a cost basis. LCC, CAPEX and OPEX were all found to be lower for the *T2* (MF/AOP) scheme and decreases for both trains with the plant capacity. OPEX was found to provide the largest contribution to the LCC (> 80%).

Process acquisition (the RO and MF processes for *T1* MF and AOP for *T2*) was found to provide the largest contribution to the CAPEX. Regarding the OPEX, labour costs were found to have a significant impact at lower plant capacity while, due to the economy of scale, the process operating costs contributed most significantly to OPEX for the larger plants. As expected for the OPEX the RO process, with its high energy, chemical and membrane replacement costs, provided the largest component of the OPEX.

A sensitivity analysis was performed to assess the effect of operating parameters on the CAPEX and the OPEX for *T1*. The RO flux was found to have the greatest impact on the CAPEX, which increased with decreasing flux due to the larger number of modules required. For the OPEX, an increase in RO

recovery lead to an increased OPEX, due to the higher operating pressure associated with higher recoveries.

8.5 References

BS ISO 14040:2006, Environmental management - Life cycle assessment - Principles and framework.

BS ISO 25686-5:2008, Buildings and constructed assets - Service-life planning - Part 5: Life-cycle costing.

Cazurra, T. (2008), Water reuse of south Barcelona's wastewater reclamation plant, *Desalination*, 218, p. 43-51.

Côté, P., Masini, M., Mourato, D. (2004), Comparison of membrane options for water reuse and reclamation, *Desalination*, 167, p. 1-11.

Hatt, J. W., Judd, S. J. and Germain, E. (2011), Screening optimisation for indirect potable reuse, *Water science and Technology*, 63 (12), p. 2846-2852.

James, C., Germain, E., Raffin, M., Tupper, M., Pearce, P. (2011), Advanced Oxidation (UV/H202) for Indirect Potable Reuse – Micro-pollutants Treatment Efficacy, In: *8th IWA International Conference on Water Reclamation & Reuse*, 26-29 September 2011, Barcelona, Spain.

Juang, L-C., Tseng, D-H., Lin, H-Y. (2006), Membrane processes for water reuse from the effluent of industrial park wastewater treatment plant: a study on flux and fouling of membrane, *Desalination*, 202, p. 302-309.

Mackey, E.D., Cushing, R.S., Crozes, G.F. (2001), Practical aspects of UV disinfection, AWWA Research Foundation.

Majamaa, K., Aerts, P.E.M., Groot, C., Paping, L.L.M.J., Van den Broek, W., Van Agtmaal, S. (2010), Industrial water reuse with integrated membrane system increases the sustainability of the chemical manufacturing, *Desalination and Water Treatement*, 18, p. 17-23.

Markus, M.R., Deshmukh, S.S. (2010), An innovative approach to water supply – the groundwater replenishment system, In: *World Environmental and Water Ressources Congress 2010: Challenges of Change*, Providence, Rhode Island 16-20 May 2010, p. 3624-3639.

Lazarova, V., Gallego, S., Molina, V.G., Rougé, P. (2008), Problems of operation and main reasons for failure of membranes in tertiary treatment systems, *Water Science and Technology*, 11, p. 1777-1784.

Pearce, G.K. (2008), UF/MF pre-treatment to RO in seawater and wastewater reuse applications: a comparison of energy costs, Desalination, 222, p. 66-73.

Raffin, M., Germain, E., Judd, S. (2011a), Optimising operation of an integrated membrane system (IMS) — A Box–Behnken approach, *Desalination*, 273, p. 136-141.

Raffin, M., Germain, E., Judd, S. (2011b), Optimisation of MF membrane cleaning protocol in an Indirect Potable Reuse (IPR) scheme, *Separation and Purification Technology*, 80, p. 452-458.

Tangsubkul, N., Beavis, P., Moore, S.J., Lundie, S., Waite, T.D. (2005), Life cycle assessment of water recycling technology, *Water Ressources Management*, 19, p. 521-537.

Wilf, M. (2010), *The guidebook to membrane technology for wastewater reclamation*, Balaban Desalination Publications

Won, W., Shields, P., Comparative life cycle costs for operation of full-scale conventional pretreament/RO and MF/RO systems, In: *AWWA Membrane Technology conference*, Long Beach, CA, 1999.

CHAPTER 9: CONCLUSIONS AND SUGGESTIONS FOR FURTHER WORK

9.1 Conclusions

Understanding of integrated membrane system (IMS) sustainability for wastewater reuse in terms of fouling minimisation and cost has been extended through practical study and modelling of a pilot scale plant fed with real secondary municipal wastewater. Statistical experimental programming, and specifically Box-Behnken design (BBD) has been successfully applied to optimise aspect of the membrane processes. From the research, the following conclusions can be drawn:

1. A literature survey and a review of nine membrane-based municipal wastewater reuse plant has revealed the extent of IMS implementation worldwide (Chapter 2). The plants differ primarily with respect to membrane operation and maintenance conditions, pre-treatment of the MF/UF-RO system being mostly screening. Across nine plants studied the MF/UF backwash interval was found to be more dependent on the feed water temperature than any other specific water quality determinants, while chemical cleaning interval was dependent on plant operating parameters such as flux and feed water quality. The colloid content of the RO feedwater, as reflected on the silt density index (SDI) and the turbidity, was found to correlate with the cleaning frequency of the RO membranes and influence other RO process operating parameters like flux and recovery. The turbidity of MF/UF filtrate roughly correlated with the membrane pore size indicating the greater rejection afforded by the smaller membrane pore size. The overall reported energy demand ranged from 0.8 to 2.3 kWh/m^3, and appeared to correlate with the MF/UF flux and, more approximately, the mathematical product of the flux, recovery and total dissolved solids in the case of the RO.

2. An evaluation of the peer-reviewed literature for statistical experimental programming methods in water and wastewater treatment in general, and membrane processing in particular, revealed five methods of which

the most popular was factorial design (Chapter 3). A comparison of Box-Behnken design (BBD) with the four other methods used revealed BBD to offer an appropriate and efficient method for experimental design for optimising membrane processes on the basis of: a) the low number of experiments required; b) the non-linear/versatile nature of the model derived from the design; c) the ability to account for interaction between parameters; d) the assessment of experimental errors through the use of central points, and e) the uniformity of the investigation within the range of parameter values studied by virtue of the equidistance between them. In the case of 4 parameters and 3 levels of parametric values, the number of tests required is 3 times lower than for a 3^n factorial design. Notwithstanding this, the method is not widely used in water and wastewater process optimisation, with only less than 100 previous literature publications from the past 10 years.

3. The application of BBD to both MF and RO membranes processes has been successfully demonstrated from pilot-scale studies. A first optimisation study defined the envelope of operating parameters of both the MF and the RO process in terms of fouling minimisation (Chapter 4) while in a second optimisation study allow to defined the operating parameter to enhance the MF cleaning in place (Chapter 6). Both of this optimisation studies demonstrate BBD to be an appropriate statistical tool for this duty, allowing a reduced number of trials to identify the optimum operating conditions. However, the results are dependent on the range of the parameters studied and cannot be extrapolated to regions outside those studied conditions.

4. The envelope of operating conditions for the MF (Chapter 4) was found to be similar to full scale operating conditions applied for such systems (Chapter 2). Lower fluxes and higher backwash frequencies reduced MF membrane fouling, corroborated by a subsequent study (Chapter 5) of

the influence of operating parameters and water quality on fouling rates. Reversible fouling was found to increase exponentially with turbidity and to follow a power or exponential relationship with flux. Irreversible fouling was found to be promoted only by increased flux and backwash interval, while reversible fouling rate depended on flux, turbidity and temperature. The Newtonian viscosity correction was shown to be insufficient to account for the influence of temperature on reversible fouling rate. This is in accordance with the findings reported in Chapter 2 where the backwash interval, which removes reversible fouling, was found to be linearly dependent on the temperature. Some residual fouling, following the same exponential or power relationship with the flux as that manifested at different turbidities, was observed at zero turbidity. Operation above the classical critical flux was found to be sustainable under appropriate backflushing conditions. It was concluded that the sustainable flux concept was a more appropriate basis for process control and optimisation than critical flux, corroborating observations made for membrane bioreactors, since critical flux takes no account of process economics. Chloramine was found to have no significant influence on short-term fouling of the MF process.

5. The impact of different chemical cleaning protocols on permeability recovery has been quantified with respect to reagent type, concentration, temperature and soaking time on a bench-scale by providing predictive model equations for the MF process (Chapter 6). Results are in agreement with those from previous studies, in that synergistic effects appear important in determining optimum chemical cleaning and the relative influence of the three key parameters is system dependent. Neither the effect nor the significance of parameters (temperature, concentration and soaking time) follow the same behaviour for each chemical reagent, again corroborating previously reported findings. This implies that the CIP must be optimised for each installation and

application, which often takes place in practice only on an inefficient, ad hoc basis. Sodium hypochlorite adjusted to pH 10 was found to be the most effective cleaning reagent, yielding a permeability recovery of 100% or more. Bench-scale data could be replicated at the pilot plant scale only if the two sets of data referred to the same operating temperature, again reflecting the inadequacy of the viscosity correction.

6. For the RO process, the envelope of operating conditions has been found to be more conservative than that usually applied to such systems, reflecting a lower quality feedwater with respect to phosphorus concentration and temperature (Chapter 4). As expected, lower pH and recovery values reduced RO membrane fouling. In this study it was found that there were no significant interactions between pH and the recovery for the range of parameters studied for the RO process, whereas synergy between backwash frequency and flux was apparent for the MF process. In this study, the range chosen for the antiscalant dosing was too low, such that the results showed no impact of this reagent.

7. Whilst over the narrow range of antiscalant dose studied in Chapter 4 no influence on RO membrane scaling was determined, this is clearly counterintuitive, since antiscalant dosing is pivotal in sustaining RO operation. Results of a study of a range of antiscalants, compared on the basis of scaling mitigation (Chapter 7), showed differing efficiencies with respect to prevent scaling. Tests performed on a future generation of antiscalants showed them to be the most effective in scaling minimisation. However, none of them appeared capable of avoiding scaling without the addition of sulphuric acid, possibly reflecting the relative inefficacy of such antiscalants against calcium phosphate colloidal fouling. A cost analysis quantified the benefit of employing a more effective antiscalant at more neutral pH levels. A small increase in

adjusted pH was shown significantly reduce operational costs due to the significant reduction in acid consumption: an operating cost reduction of up to 77% can be obtained by increasing adjusted pH from 6.25 to 7.25 at a wastewater alkalinity of 195 mg/L as $CaCO_3$. Capital costs would also be reduced resulting from the marginally fewer membrane elements required at longer chemical cleaning intervals and commensurately higher net fluxes.

8. Membrane autopsies conducted to assess fouling of the RO membranes revealed the first and the second stages of the array to be subject to significantly less scaling than the third stage (Chapter 7). Scaling was mostly associated with calcium phosphate, although calcium carbonate was also present. Biofouling was observed on the three stages with higher concentrations at the 3rd stage. This was attributed to a considerably depleted chloramine residual, whose concentration was shown to decrease from 1 mg/L to <0.1 mg/L across the array.

9. A life cycle cost (LCC) analysis of two treatment trains (MF/RO/AOP and MF/AOP) at three plant capacities revealed a lower CAPEX and OPEX, and thus a correspondingly lower LCC, for the MF/AOP scheme (Chapter 8). LCC decreases for both schemes with increasing plant capacity. CAPEX mainly derives from process acquisition, as opposed to CAPEX from the building or electrical and pipe work, while the primary contribution to the OPEX varies with capacity; the highest contribution to process OPEX at small scale is labour, whilst at large scale it is the process. Sensitivity analysis showed the RO flux to exert the greatest influence on CAPEX, since fewer RO membrane elements are required at higher fluxes. RO recovery significantly affects OPEX due to its impact on energy consumption.

9.2 Suggestions for further work

The work reported has focused almost entirely on the sustaining of membrane permeability and analysing costs. The necessary assumption has also been made that over the period of the experiment performed the change in water quality, as reflected primarily by aggregate parameters such as TOC, turbidity and TDS, does not change. It has also been asserted that BBD is most appropriate for optimisation. Finally, it is assumed that membrane technology offers the best solution for indirect potable reuse. All of these assumptions ultimately are either questionable to a greater or lesser extent or otherwise constrain the interpretation of the results.

It is also the case that, whilst fouling control and cost are pivotal considerations in membrane processes, the issue of micropollutants – substances considered hazardous to the environmental and/or human health even at comparatively very low concentrations – has become increasingly important in the water industry. This topic is particularly germane to membrane processes since reverse osmosis is arguably the only water/wastewater treatment process providing an effective absolute barrier to some of the more recalcitrant micropollutants, and toxic metal ions in particular.

It would therefore be beneficial to concentrate further work in three areas, as indicated below:

1. The assessment of the impact of other water quality determinants on plant operation, and phosphate in particular. Evidence from the study suggests that there may be a significant influence exerted on the antiscalant demand and/or efficacy, as a function of pH, by the phosphate concentration.

2. The assessment of an alternative statistical experimental design should be conducted for comparative purposes. Whilst BBD appears to offer many advantages and has been successfully demonstrated, it must be

acknowledged that by far the most well used method reported in the literature is factorial design.

3. The assessment of the overall environmental impact of the process, rather than simply cost. Given the increasing emphasis on carbon, reflected by the UK's commitment to reduce is carbon emissions by 60% below the 1990 level by 2050, a carbon footprint assessment of the plant over its life cycle is more appropriate than a simple cost analysis. Life cycle analyses of membrane processes can be found in literature but few have focused on a complete treatment train.

4. The fate of micropollutants, and in particular those not readily removed by conventional biological or wastewater polishing processes (primarily filtration or adsorption) should be assessed. This should include a consideration of the management of the concentrate stream, since this will contain the rejected micropollutants which may then need to be removed from this stream by classical means (coagulation-clarification, adsorption, etc).T

5. The efficacy of other technologies should be assessed with a view to displacing the RO process, which has a high energy demand and generates a potentially problematic concentrate stream. Whilst many wastewater reuse schemes exist which are based on the two-stage MF/UF-RO process, it should not be assumed that this represents the most sustainable option for the future.

REFERENCES, complete

Alhadidi, A., Kennedy, M., Diepeven, A., Prummel, H., Boorsma, M., Schippers, J.C. (2009), Scaling potential calculations using different methods, *Desalination and Water Treatment*, 6, p. 138-143.

Amin, I.N.H.M., Mohammad, A.W., Markom, M., Peng, L.C., Hilal, N. (2010), Flux decline study during ultrafiltration of glycerine-rich fatty acid solutions, *Journal of Membrane Science*, 351, p. 75-86.

Asano, T. (2006), *Water reuse: issues, technologies, and applications*, Metcalf and Eddy, 1st Ed., McGrawHill, New York.

Bacchin, P. (2004), A possible link between critical and limiting flux for colloidal systems: consideration of critical deposit formation along a membrane, *Journal of Membrane Science*, 228, p. 237-241.

Bacchin, P., Aimar, P. (2005), Critical fouling conditions induced by colloidal surface interaction: from causes to consequences, *Desalination*, 175, p. 21-27.

Bacchin, P., Aimar, P., Field, R.W. (2006), Critical and sustainable fluxes: Theory, experiments and applications, *Journal of Membrane Science*, 281, p. 42-69.

Bartels C.R., Franks R., Furukawa R., Murkute P., Papukchiev U. (2004), Integrated membrane system for low fouling RO desalting of municipal wastewater, final report for the Desalination Research and Innovation Partnership, conducted by the City of Oceanside in cooperation with Hydranautics.

Bartels C.R., Wilf M., Andes K. Iong J. (2005), Design considerations for wastewater treatment by reverse osmosis, *Water Science & Technology*, 51 (6-7), p. 473-482.

Bartels, C., Wilf, M., Andes, K., Long, J. (2007), Design considerations for wastewater treatment by reverse osmosis, *Water and Wastewater Asia*, May/June 2007, p. 20-26.

Bartlett, M., Bird, M. R. and Howell, J. A. (1995), An experimental study for the development of a qualitative membrane cleaning model, *Journal of Membrane Science,* 105 (1-2), p. 147-157.

Bartolomé, L., Lezamiz, J., Etxebarria, N., Zuloaga, O. and Jönsson, J. A. (2007), Determination of trace levels of dinitrophenolic compounds by microporous membrane liquid-liquid extraction in environmental water samples, *Journal of Separation Science,* 30 (13), p. 2144-2152.

Besseris, G. J. (2008), Multi-response optimisation using Taguchi method and super ranking concept, *Journal of Manufacturing Technology Management,* 9 (8), p. 1015-1029.

Bessiere, Y., Abidine, N., Bacchin, P. (2005), Low fouling condition in dead-end filtration: Evidence for a critical filtered volume and interpretation using critical osmotic pressure, *Journal of Membrane Science*, 264, p. 37-47.

Bezerra, M. A., Santelli, R. E., Oliveira, E. P., Villar, L. S. and Escaleira, L. A. (2008), Response surface methodology (RSM) as a tool for optimization in analytical chemistry, *Talanta,* 76 (5), p. 965-977.

Birima, A. H., Mohd Noor, M. J. M., Mohammed, T. A., Idris, A., Muyibi, S. A., Nagaoka, H., Ahmed, J. and Ghani, L. A. A. (2009), The effects of SRT, OLR and feed temperature on the performance of membrane bioreactor treating high strength municipal wastewater, *Desalination and Water Treatment,* 7 (1-3), p. 275-284.

Box G.E.P. and Behnken D.W. (1960), Some new three level designs for the study of quantitative variables, *Technometrics*, 2 (4), 455-475.

Box, G.P., Drapaer, N.R. (1987), *Empirical Model-Building and Response surfaces*, John Wily and Sons, Inc., USA, p 477.

Brepols, C. (2011), *Operating large scale membrane bioreactors for municipal wastewater treatment*, IWA Publishing, 1st Ed., London, UK.

Bohner, H. F. and Bradley, R. L. (1990), Effective Control of Microbial Populations in Polysulfone Ultrafiltration Membrane System, *Journal of Dairy Science,* 73 (9), p. 2309-2317.

Bourgeous, K.N., Darby, J.L., Tchobanoglous, G. (2001), Ultrafiltration of wastewater: effect of particles, mode of operation, and backwash effectiveness, *Water Research*, 35 (1), p. 77-90.

Brant, J. A., Kwan, P., Daniel, U. and Valencia, R. (2010), Pilot-scale evaluation of chemical cleaning protocols for organic and biologically fouled microfiltration membranes, *Journal of Environmental Engineering,* 136 (5), p. 542-553.

Brookes, A., Jefferson, B., Guglielmi, G., Judd, S.J. (2006), Sustainable flux fouling in a membrane bioreactor: Impact of flux and MLSS, *Separation Science and Technology*, 41, p. 1279-1291.

BS ISO 14040:2006, Environmental management - Life cycle assessment - Principles and framework.

BS ISO 25686-5:2008, Buildings and constructed assets - Service-life planning - Part 5: Life-cycle costing.

Cabassud M., Delgrange-Vincent N., Cabassud C., Durand-Bourlier L., Lainé J.M. (2002), Neural networks: a tool to improve UF plant productivity, *Desalination*, 145, 223-231.

Carić, M.D., Milanović, S.D., Krstić, D.M., Tekić, M.N. (2000), Fouling of inorganic membranes by adsorption of whey proteins, *Journal of Membrane Science*, 135, p. 83-88.

Cazurra, T. (2008), Water reuse of south Barcelona's wastewater reclamation plant, *Desalination*, 218, p. 43-51.

Chen, J.P., Kim, S.L., Ting, Y.P. (2003), Optimization of membrane physical and chemical cleaning by a statistically designed approach, *Journal of membrane Science*, 219, p. 27-45.

Cho, J., Amy, G., Pellegrino, J. (2000), Membrane filtration of natural organic matter: factors and mechanisms affecting rejection and flux decline with charged ultrafiltration (UF) membrane, *Journal of Membrane Science*, 164, p. 89-110.

Citulski, J.A., Farahbakhsh, K., Kent, F.C. (2009), Effects of total suspended solids loading on short-term fouling in the treatment of secondary effluent by an immersed ultrafiltration pilot system, *Water Environment Research*, 81, p. 2427-2436.

Côté, P., Masini, M., Mourato, D. (2004), Comparison of membrane options for water reuse and reclamation, *Desalination*, 167, p. 1-11.

Daramola, M.O., Keesman, K.J., Spenkelink, F. (2007), Process modelling of ultrafiltration units: A RSM approach, *Journal of Applied Sciences*, 7 (23), 3687-3695.

Darcovich, K., Dal-Cin, M. M., Ballèvre, S. and Wavelet, J. (1997), CFD-assisted thin channel membrane characterization module design, *Journal of Membrane Science,* 124 (2), p. 181-193.

Du Pisani, P. L. (2006), Direct reclamation of potable water at Windhoek's Goreangab reclamation plant, *Desalination,* 188 (1-3), p. 79-88.

Esteban R. I., Ortega de Miguel, E. (2008), Present and future of wastewater reuse in Spain, *Desalination*, 218, p. 105-119.

Ferreira, S. L. C., Bruns, R. E., Ferreira, H. S., Matos, G. D., David, J. M., Brandão, G. C., da Silva, E. G. P., Portugal, L. A., dos Reis, P. S., Souza, A. S. and dos Santos, W. N. L. (2007), Box-Behnken design: An alternative for the optimization of analytical methods, *Analytica Chimica Acta,* 597 (2), p. 179-186.

Figueroa, R. A., Cassano, A. and Drioli, E. (2011), Ultrafiltration of orange press liquor: Optimization for permeate flux and fouling index by response surface methodology, *Separation and Purification Technology,* 80 (1), p. 1-10.

Freeman, S., Bates, J., Wallis-Lage, C., McEvoy, J. (2008), Drought relief in South East, Queensland, Australia, provided by membrane-reclaimed water, *AWWA Journal*, 100 (2), p. 40-52.

Gabelich, C. J., Ishida, K.P., Gerringer, W., Evangelista, R., Kalyan, M., Suffet, I.H. (2006), Control of residual aluminium from conventional treatment to improve reverse osmosis performance, *Desalination*, 190, p. 147-160.

Ghafour E.E.A. (2002), Enhancing RO system performance utilizing antiscalant, *Desalination*, 153, 149-153.

Gómez Gotor, A., Pérez Baez, S.O., Espinoza, C.A., Bachir, S.I. (2001), Membrane processes for the recovery and reuse of wastewater in agriculture, *Desalination*, 137, p. 187-192.

Greenberg, G., Hasson, D., and Semiat, R. (2005). Limits of RO recovery imposed by calcium phosphate precipitation, *Desalination*, 183, p. 273-288.

Goldman G., Starosvetsky J., Armon R. (2009), Inhibition of biofilm formation on UF membrane by use of specific bacteriophages, *Journal of Membrane Science*, 342, p. 145-152.

Gönder, Z. B., Kaya, Y., Vergili, I. and Barlas, H. (2010), Optimization of filtration conditions for CIP wastewater treatment by nanofiltration process using Taguchi approach, *Separation and Purification Technology,* 70 (3), p. 265-273.

Hatt, J., Germain, E., and Judd, S.J. (2011). Screening optimisation for indirect potable reuse, *Water Science and Technology*, 63 (12), p. 2846-2852.

Heijman, S., Vantighem, M., Raktoe, S., Verberk, J., van Dijk, J. (2007), Blocking of capillaries as fouling mechanism for dead-end ultrafiltration, *Journal of Membrane Science*, 287, p. 119-125

Hesampour, M., Krzyzaniak, A. and Nyström, M. (2008), The influence of different factors on the stability and ultrafiltration of emulsified oil in water, *Journal of Membrane Science,* 325 (1), p. 199-208.

Hills, S., Birks, R., Grant, E., Aitken, V. (2007), Feasibility studies of planned indirect potable reuse for augmenting future water supplies in London, In: *Wastewater Reclamation and Reuse for Sustainability 2007 Conference*, Antwerp, Belgium, 9-11 March 2007.

Hinkelmann, K. and Jo, J. (1998), Linear trend-free Box-Behnken designs, *Journal of Statistical Planning and Inference,* 72 (1-2), p. 347-354.

Hossam-Eldin, A., El-Nashar, A. M. and Ismaiel, A. (2009), Techno-economic optimization of SWRO desalination using advanced control approaches, *Desalination and Water Treatment,* 12 (1-3), p. 389-399.

Huang, H., Young, T., Jacangelo, J. G. (2009), Novel approach for the analysis of bench-scale, low pressure membrane fouling in water treatment, *Journal of Membrane Science,* 334 (1-2), p. 1-8.

Hwang, K.-J., Chan, C.-S., Tung, K.-L. (2009), Effect of backwash on the performance of submerged membrane filtration, *Journal of Membrane Science*, 330, p. 349-356.

James, C., Germain, E., Raffin, M., Tupper, M., Pearce, P. (2011), Advanced Oxidation (UV/H202) for Indirect Potable Reuse – Micro-pollutants Treatment Efficacy, In: *8th IWA International Conference on Water Reclamation & Reuse*, 26-29 September 2011, Barcelona, Spain.

Jarsutthirak, C., Amy, G., Croué, J-P. (2002), Fouling characteristics of wastewater effluent organic matter (EfOM) isolates on NF and UF membranes, *Desalination*, 145, p. 247-255.

Jiang, T., Kennedy, M.D., Guinzbourg, B.F., Vanrolleghem, P.A., Schippers, J.C. (2005), Optimising the operation of a MBR pilot plant by quantitative

analysis of the membrane fouling mechanism, *Water Science and Technology*, 51 (6-7), p. 19-25.

Jimenez, B., Asano T. (2008), *Water Reuse, an international survey of current practice, issues and needs*, 1st Ed., IWA Publishing, London, UK.

Jokic, A., Zavargo, Z., Seres, Z., Tekic, M. (2010), The effect of turbulence promoter on cross-flow microfiltration of yeast suspensions: A response surface methodology approach, *Journal of Membrane Science*, 350, p. 269-278

Juang, L-C., Tseng, D-H., Lin, H-Y. (2006), Membrane processes for water reuse from the effluent of industrial park wastewater treatment plant: a study on flux and fouling of membrane, *Desalination*, 202, p. 302-309.

Judd, S. (2010), *The MBR book: principles and applications of membrane bioreactors in water and wastewater treatment*, 2nd Ed., Elsevier, Oxford, UK.

Khayet M., Cojocaru C., Zakrzewska-Trznadel G. (2008), Response surface modelling and optimisation in pervaporation, *Journal of Membrane Science*, 321, p. 272-283.

Kim, J., DiGiano, F.A., Reardon, R.D. (2008), Autopsy of high-pressure membranes to compare effectiveness of MF and UF pre-treatment in water reclamation, *Water Research*, 42, p. 697-706.

Kimura, K., Yamamura, H., Watanabe, Y. (2006), Irreversible fouling in MF/UF membranes caused by Natural Organic Matters (NOMs) isolated from different origins, *Separation Science and Technology,* 41 (7), p. 1331-1344.

Lai, W-L., Chen, L-F., Chen, J-J., Liao, S-W. (2009), Effects of operational parameters on carobon recovery and water flux in ultrafiltration using fractional factorial design, *Desalination*, 249, p. 1365-1370.

Latinen N. (2002), Development of a ceramic membrane filtration equipment and its applicability for different wastewaters. PhD thesis, Lappeenranta University of Technology.

Lazarova, V., Shields, P., Levine, B., Savoye, P., Hranisavljevic, D., Renaud, P. 92003), Production of high quality for reuse purposes: the West Basin experience, *Water Science and Technology: Water Supply*, 3 (3), p. 167-175.

Le-Clech, P., Jefferson, B., Chang, I.S., Judd, S.J. (2003), Critical flux determination by the flux-step method in a submerged membrane bioreactor, *Journal of Membrane Science*, 227, p. 81-93.

Le-Clech, P., Chen, V., Fane, T.A.G. (2006), Fouling in membrane bioreactors used in wastewater treatment, *Journal of Membrane Science*, 284, p. 17-53.

Lee, N., G. Amy, J-P. Croué J-P., Buisson, H. (2004), Identification and understanding of fouling in low-pressure membrane (MF/UF) filtration by natural organic matter (NOM), *Water Research*, 38, p. 4511-4523.

Lee Y-G., Gambierb A., Badreddinb E., Leec S., Yangd D.R., Kim J.H. (2009), Application of hybrid systems techniques for cleaning and replacement of a RO membrane, *Desalination*, 247, p. 25-32.

Li, X., Li, J., Fu, X., Wickramasinghe, R. and Chen, J. (2005), Chemical cleaning of PS ultrafilters fouled by the fermentation broth of glutamic acid, *Separation and Purification Technology,* 42 (2), p. 181-187.

Li, Y., Zhang, W., Zhang, X., Chen, C., Wang, J. (2010), Characterisation of fouling in immersed polyvinylidene fluoride hollow fibre membrane ultrafiltration by particles and natural organic matter, *Desalination and Water Treatment*, 18, p. 309-314.

Libotean, D., Giralt, J., Giralt, F., Rallo, R., Wolfe, T., Cohen, Y. (2009), Neural Network approach for modelling the performance of reverse osmosis membrane desalting, *Journal of Membrane Science*, 326, p. 408-419.

Lin, S-H., Hung, C-L., Juang R-S. (2008), Effect of operating parameters on the separation of proteins in aqueous solutions by dead-end ultrafiltration, *Desalination*, 234, p. 116-125

Lin H. and Bérubé P.R. (2007), Modelling the impact of permeate flux and hydrodynamic conditions on fouling in submerged hollow fiber membranes, *Water Science & Technology: Water Supply*, 7(4), p. 111-118.

Liu, Q-F., Kim, S-H., Lee, S. (2009), Prediction of microfiltration membrane fouling using artificial neural network models, *Separation and Purification Technology*, 70, p. 96-102.

Lodge, B.N., Judd, S.J., and Smith, A.J. (2001), A statistical method for quantifying the different fouling effects of three combined water sources on an ultrafiltration membrane, *Desalination*, 142, p. 143-149 (2001).

Lozier, J. (2000), Two approaches to indirect potable reuse using membrane technology, *Water Science and Technology,* 41 (10-11), p. 149-156.

MacBeth, I., Murrer, J., Latter, S. (2004), Reducing the costs for ultra-pure water production for use as de-min plant feed water for Peterborough power station, UK, Chemie im Kraftwerk 2004.

Mackey, E.D., Cushing, R.S., Crozes, G.F. (2001), Practical aspects of UV disinfection, AWWA Research Foundation.

Majamaa, K., Aerts, P.E.M., Groot, C., Paping, L.L.M.J., Van den Broek, W., Van Agtmaal, S. (2010), Industrial water reuse with integrated membrane system increases the sustainability of the chemical manufacturing, *Desalination and Water Treatement*, 18, p. 17-23.

McAdam, E.J., Judd, S.J. (2008), Optimisation of dead-end filtration for an immersed anoxic membrane bioreactor, *Journal of Membrane Science*, 325, p. 940-946.

McCullagh P. and Nelder J.A. (1989), *Generalized linear model*, 2nd Ed., Chapman & Hall, 13-14.

Markus, M.R., Deshmukh, S.S. (2010), An innovative approach to water supply - The Groundwater Replenishment System, In: *World Environmental and Water*

Resources Congress 2010: Challenges of Change, Providence, Rhode Island 16-20 May 2010, p. 3624-3639.

Mendoza-Roca, J., Galiana-Aleixandre, M., Lora-Garcia, J., Bes-Pia, A. (2010), Purification of tannery effluents by ultrafiltration in view of permeate reuse, *Separation and Purification Technology*, 70, p. 296-301

Meyers, R.H., Montgomery, D.C., Anderson-Cook, C.M. (2009), *Response surface methodology – Process and product optimization using designed experiments*, 3rd Edition, Whiley, New York.

Mohammadi, T., Madaeni, S. S. and Moghadam, M. K. (2003), Investigation of membrane fouling,, *Desalination,* 153 (1-3), p. 155-160.

Moon, J., Kang, M-S., Lim, J-L., Kim, C-H., Park, H-D. (2009), Evaluation of a low-pressure membrane filtration for drinking water treatment: pre-treatment by coagulation/sedimentation for the MF membrane, *Desalination*, 247(1-3), p. 271-284.

Mosteo, R., Ormad, P., Mozas, E., Sarasa, J. and Ovelleiro, J. L. (2006), Factorial experimental design of winery wastewaters treatment by heterogeneous photo-Fenton process, *Water research,* 40 (8), p. 1561-1568.

Mousa, H.A., Al-Hitmi, S.A. (2007), Treatability of wastewater and membrane fouling. *Desalination*, 217, p. 65-73.

Nair, V. N., Abraham, B., MacKay, J., Nelder, J. A., Box, G., Phadke, M. S., Kacker, R. S., Sacks, J., Welch, W. J., Lorenzen, T. J., Shoemaker, A. C., Tsui, K. L., Lucas, J. M., Taguchi, S., Myers, R. H., Vining, G. G. and Wu, C. F. J. (1992), "Taguchi´s Parameter Design: A panel discussion ", *Technometrics,* 34 (2), p. 127-161.

Nguyen, T., Fan, L., Roddick, F.A., Harris, J.L. (2009), A comparative study of microfiltration and ultrafiltration of activated sludge-lagoon effluent, *Desalination*, 236, p. 208-215.

Ning, R.Y., Troyer, T.L. (2007), Colloidal fouling of RO membranes following MF/UF in the reclamation of municipal wastewater, *Desalination*, 208, p. 232-237.

Ognier, S., Wisniewski, C., Grasmick, A., Membrane fouling during constant flux filtration in membrane bioreactors, *Membrane Technology July 2002*.

Onsekizoglu, P., Savas Bahceci, K. and Acar, J. (2010), The use of factorial design for modeling membrane distillation, *Journal of Membrane Science,* 349 (1-2), p. 225-230.

Parameshwaran, K., Fane, A.G., Cho, B.D., Kim, K.J. (2001), Analysis of microfiltration performance with constant flux processing of secondary effluent, Water Research, 35 (18), p. 4349-4358.

Park, C., Hong, S-W., Chung, T.H., Choi, Y-S. (2010), Performance evaluation of pre-treatment processes in integrated membrane system for wastewater reuse, *Desalination*, 250, p. 673-676.

Pearce, G.K. (2008), UF/MF pre-treatment to RO in seawater and wastewater reuse applications: a comparison of energy costs, Desalination, 222, p. 66-73.

Peng, W.. Escobar, I.C., White, D.B. (2004), Effect of water chemistries and properties of membrane on the performance and fouling - a model development study, *Journal of Membrane Science*, 238 (1-2), p. 33-46.

Pointié, M., Rapenne, S., Thekkedath, A., Duchesne, J., Jacquemet, V., Leparc, J., Suty, H. (2005), Tools for membrane autopsies and antifouling strategies in seewater feeds: a review, *Desalination*, 181, p. 75-90.

Popovic, S., Milanovic, S., Ilicic, M., Djuric, M., Tekic, M. (2009), Flux recovery of tubular ceramic membranes fouled with whey proteins, *Desalination*, 249, p. 293-300

Porcelli, N., Hillis, P. and Judd, S. (2009), Microfiltration membrane plant start up: A case study with autopsy and permeability recovery analysis, *Environmental Technology*, 30 (6), p. 629-639.

Porcelli, N. and Judd, S. (2010), Chemical cleaning of potable water membranes: A review, *Separation and Purification Technology,* 71 (2), p. 137-143.

Porcelli, N. and Judd, S. (2010), Chemical cleaning of potable water membranes: The cost benefit of optimisation, *Water research,* 44 (5), p. 1389-1398.

Porcelli, N. and Judd, S. (2010), Impact of cleaning protocol on membrane permeability recovery: a sensitivity analysis, *J. AWWAF*, 102 (12)

Psoch, C., Shiewer, S. (2006), Resistance analysis for enhanced wastewater membrane filtration, *Journal of Membrane Science*, 280, p. 284-297.

Raffin. M., Germain, E., Judd, S. (2011), Optimising operation of an integrated membrane system (IMS) — A Box–Behnken approach, *Desalination*, 273, p. 136-141.

Raffin, M., Germain, E., Judd, S. (2011), Optimisation of MF membrane cleaning protocol in an Indirect Potable Reuse (IPR) scheme, *Separation and Purification Technology*, 80, p. 452-458.

Rajasimman, M., Sangeetha, R. and Karthik, P. (2009), Statistical optimization of process parameters for the extraction of chromium(VI) from pharmaceutical wastewater by emulsion liquid membrane, *Chemical Engineering Journal,* 150 (2-3), p. 275-279.

Rodriguez, D., Van Buynder, P., Lugg, R., Blair, P., Devine, B., Cook, A. and Weinstein, P. (2009), Indirect potable reuse: A sustainable water supply alternative, *International Journal of Environmental Research and Public Health,* 6 (3), p. 1174-1209.

Roux, A., Robillot, C., Chapman, H., Leusch, F., Hodge, M., Walker, T. (2010), Hazard identification, qualitative risk assessment and monitoring on the Western Corridor Recycled Water Project, In: *7th IWA Specialty Conference on Wastewater Reclamation & Reuse, 21-25 September 2009, Brisbane, Australia*.

Sadeddin, K., Naser, A., Firas, A. (2011), Removal of turbidity and suspended solids by electro-coagulation to improve feed water quality of reverse osmosis plant, *Desalination*, 268, p. 204-207.

Saleh, J., Dubé, M. A. and Tremblay, A. Y. (2010), "Effect of soap, methanol, and water on glycerol particle size in biodiesel purification", *Energy and Fuels,* 24 (11), p. 6179-6186.

Seah, H., Tan, T.P., Chong, M.L. and Leong, J., (2008), NEWater-multi safety barrier approach for indirect potable use. Water Science & Technology: Water Supply, 8 (5), p. 573–588.

Sauvet-Goichon B. (2007), Ashkelton desalination plant – a successful challenge, *Desalination*, 203, p. 75-81.

She, Q., Tang, C.Y., Wang, Y-N., Zhang, Z. (2009), The role of hydrodynamic conditions and solution chemistry on protein fouling during ultrafiltration, *Desalination*, 249, p. 1079-1087.

Sivakumar, M., Annadurai, G., Mohan, D. (1999), Studies on Box-Behnken design experiments : cellulose acetate-polyurethane ultrafiltration membranes for BSA separation, *Bioprocess Engineering*, 21, p. 65-68.

Soice, N.P., Maladono, A.C., Takigawa, D.Y., Norman, A.D., Krantz, W.B., Greenberg, A.R. (2003), Oxidative degradation of polyamide RO membranes: studies of molecular model compounds and selected membranes, *Journal of Applied Polymer Science*, 90 (5), p. 1173-1184.

Strugholtz, S., Sundaramoorthy, K., Panglisch, S., Lerch, A., Brügger, A. and Gimbel, R. (2005), Evaluation of the performance of different chemicals for cleaning capillary membranes, *Desalination,* 179 (1-3), p. 191-202.

Tang X., Flint, S., Benett, S., Brooks, J. (2010), The efficacy of different cleaners and sanatisers in cleaning biofilms on UF membranes used in the dairy industry, *Journal of Membrane Science*, 352, p. 71-75

Tangsubkul, N., Beavis, P., Moore, S.J., Lundie, S., Waite, T.D. (2005), Life cycle assessment of water recycling technology, *Water Ressources Management*, 19, p. 521-537.

Tam, L.S., Tang, T.W., Lau, G.N., Sharma, K.R., Chen, G.H. (2007), A pilot study for wastewater reclamation and reuse with MBR/RO and MF/RO systems, *Desalination*, 202, p. 106-113.

Tansel, B., Regula, J., Shalewitz, R. (2000), Evaluation of ultrafiltration process performance for treatment of petroleum contaminated waters, *water, Air, and Soil Pollution*, 126, p. 291-305.

Tarley, C. R. T., Silveira, G., dos Santos, W. N. L., Matos, G. D., da Silva, E. G. P., Bezerra, M. A., Miró, M. and Ferreira, S. L. C. (2009), Chemometric tools in electroanalytical chemistry: Methods for optimization based on factorial design and response surface methodology, *Microchemical Journal,* 92 (1), p. 58-67.

Tercero Espinoza L.A., Rembor M., Arriba Matesanz C., Heidt A., Frimmel F.H. (2010), Formation of bromoform in irradiated titanium dioxide suspensions with varying photocatalyst, dissolved organic carbon and bromide concentration, *Water Research*, 43, p. 4143-4148.

Tian, J., Chen, Z., Yang, Y., Liang, H., Nan, J. and Li, G. (2010), Consecutive chemical cleaning of fouled PVC membrane using NaOH and ethanol during ultrafiltration of river water, *Water Research,* 44 (1), p. 59-68.

Thompson M. and Powell D. (2003), Case study – Kranji high grade water reclamation plant, Singapore, *IMSTEC'03*, September 2003, Australia.

Tran-Ha, M. H. and Wiley, D. E. (1998), The relationship between membrane cleaning efficiency and water quality, *Journal of Membrane Science,* 145 (1), p. 99-110.

Ujang, Z., Ng. K.S., Hamzah T.H.T., Roger P, Ismail M.R., Shahabudin S.M, Abdul Hamid M.H. (2007), Application of immersed MF (IMF) followed by

reverse osmosis (RO) membrane for wastewater reclamation: A case study in Malaysia, *Water Science and Technology*, 56 (9), p. 103-108.

US EPA, Membrane Filtration guidance manual, EPA 815-R-06-009, November 2005.

Väisänen, P., Bird, M. R. and Nyström, M. (2002), Treatment of UF membranes with simple and formulated cleaning agents, *Food and Bioproducts Processing: Transactions of the Institution of of Chemical Engineers, Part C,* 80 (2), p. 98-108.

Van Houtte, E., Verbauwhede, J. (2008) Operational experience with indirect potable reuse at the Flemish Coast, *Desalination*, 218, p. 198–207.

Villacorte, L.O., Kennedy, M.D., Amy, G.L., Schippers, J.C. (2009), The fate of transparent exopolymer particles (TEP) in integrated membrane systems: Removal through pre-treatment processes and deposition on reverse osmosis membranes, Water Research, 43, p. 5039-5052.

Vrouwenvelder, J.S., Beyer, F., Dahmani, K., Hasan, N., Galjaard, G., Kruithof, J.C., Van Loosdrecht, M.C.M. (2010), Phosphate limitation to control biofouling, *Water Research*, 44, p. 3454-3466.

Vrouwenvelder, J.S., Manolaraskis, S.A., Veenendaal, H.R., Van der Kooij, D. (2000), Biofouling potential of chemicals used for scale control in RO and NF membranes, *Desalination*, p. 132, 1-10.

Wang, S-S. (1988), Effect of solution viscosity on ultrafiltration flux, *Journal of Membrane Science*, 39, p. 187-194.

Wang, L., Wang, X., Fukushi, K-I. (2008), Effects of operational conditions on ultrafiltration membrane fouling, *Desalination*, 229, p. 181-191.

Wilf, M. (2010), *The guidebook to membrane technology for wastewater reclamation*, 1st Ed., Balaban Desalination Publications, Hopkinton, USA.

Won, W., Shields, P., Comparative life cycle costs for operation of full-scale conventional pretreament/RO and MF/RO systems, In: *AWWA Membrane Technology conference*, Long Beach, CA, 1999.

Wu, J., Le-Clech, P., Stuetz, R. M., Fane, A. G. and Chen, V. (2008), "Novel filtration mode for fouling limitation in membrane bioreactors", *Water research,* 42 (14), p. 3677-3684.

Xu P., Bellona C., Drewes J.E. (2010), Fouling of nanofiltration and reverse osmosis membranes during municipal wastewater reclamation: Membrane autopsy results from pilot-scale investigations, *Journal of Membrane Science*, 353, p. 111-121.

Yamamura, H., Kimura, K., Watanabe, Y. (2007), Mechanism involved in the evolution of physically irreversible fouling in microfiltration and ultrafiltration membranes used for drinking water treatment, *Environmental Science and Technology*, 41 (19), p. 6789-6794

Yeong, Y. F., Abdullah, A. Z., Ahmad, A. L. and Bhatia, S. (2009), "Process optimization studies of p-xylene separation from binary xylene mixture over silicalite-1 membrane using response surface methodology", *Journal of Membrane Science,* 341 (1-2), p. 96-108.

Yi, X. S., Shi, W. X., Yu, S. L., Li, X. H., Sun, N. and He, C. (2011), "Factorial design applied to flux decline of anionic polyacrylamide removal from water by modified polyvinylidene fluoride ultrafiltration membranes", *Desalination,* 274 (1-3), p. 7-12.

Yu, C-H., Fang, L-C., Lateef, S., Wu C-H., Lin, C-F. (2010), Enzymatic treatment for controlling irreversible membranefouling in cross-flow humic acid-fed ultrafiltration, *Journal of Hazardous Materials*, 177, p. 1153-1158.

Zheng, X., Ernst, M., Jekel, M. (2009), Identification and quantification of major organic foulants in treated domestic wastewater affecting filterability in dead-end filtration, *Water Research*, 43, p. 238-244.

Zondervan, E. and Roffel, B. (2007), Evaluation of different cleaning agents used for cleaning ultra filtration membranes fouled by surface water, *Journal of Membrane Science,* 304 (1-2), p. 40-49.

Zularisam, A.W., Ismail, A.F., Salim, M.R., Sakinah, M., Matsuura, T. (2009), Application of coagulation-ultrafiltration hybrid process for drinking water treatment: Optimization of operating conditions using experimental design, *Separation and Purification Technology*, 65 (2), p. 193-210.

APPENDICES

APPENDIX 1: EXISTING IMS WASTEWATER REUSE PLANTS

	Country	Plant	Operation year	Capacity ($m^3.d^{-1}$)	Feed water	Pre-treatments	MF/UF	RO	Post-treatments	Applications
1	Australia	Sydney Olympic site	2000	7,500	Secondary effluent	N/A	Siemens-Memcor	N/A	N/A	Municipal non-potable reuse
2		Kwinana	2004	16,700	Secondary effluent	2 mm basket strainers NaOCl H_2SO_4	Siemens-Memcor (CMF-S)	Dow	CO_2 stripping Chlorination	Industry
3		Wollongong	2006	20,000	Tertiary effluent	N/A	Siemens-Memcor (CMF-S)	Dow	N/A	Industry
4		Bundamba	2008	36,000	Secondary effluent	Coagulation Flocculation Lamella clarifier Screening Chloramination	Siemens-Memcor	Koch	UV+ H_2O_2 Lime $CaCO_3$ Chlorine	Industry
5		Gibson Island	2008	68,000	Secondary effluent	Actiflo	Siemens-Memcor	Hydranautics	UV+ H_2O_2 Lime $CaCO_3$ Chlorine	Industry
6		Luggage Point	2008	66,000	Secondary effluent	Coagulation Flocculation Lamella clarifier	Pall (Microza UNA-620 A)	Toray (TML-10)	UV (Trojan) + H_2O_2 $CaCO_3$ NaOCl	Industry, reservoir replenishment
7	Belgium	Torreele-IWVA	2002	2,500	Secondary effluent	1 mm pre-screen Chlorination/chloramination	Zenon (ZWC500C)	Dow (BW 30LE-440)	NaOH	Indirect potable reuse via aquifer
8	China	TEDA Tianjing Economy Developping area	2003	30,000	Secondary effluent	Bacteriostasis agent	Siemens-Memcor	Toray	N/A	Non-potable municipal use and cooling tower
9	Korea	Samsung chemicals Co.	1996	30,000	N/A	N/A	Siemens-Memcor	N/A	N/A	Industry
10	Kuwait	Sulaibiya	2005	320,000	Secondary effluent	0.06 mm drumfilter Coagulation	Norit (X Flow XIGA S225 FSFC UFC M5 0.8)	Toray (TML20-400)	CO_2 stripping, NaOH, Chlorination	Industry

	Country	Plant	Operation year	Capacity ($m^3.d^{-1}$)	Feed water	Pre-treatments	MF/UF	RO	Post-treatments	Applications
11	Singapore	Bedok	Nov. 2002	32,000	Secondary effluent	0.5 mm pre-screen	Zenon (ZW500c)	Hydranautics (LFC-1)	UV (Hanovia)	Industry, reservoir replenishment
12		Kranji	Nov. 2002	40,000	Secondary effluent	0.5 mm pre-screen	Siemens-Memcor (CMF-S)	Hydranautics (LFC-1)	UV (Hanovia)	Industry, reservoir replenishment
13		Seletar	2004	24,000	Secondary effluent	N/A	Hyflux (Krystal 300B)	Toray (TML20)	UV (Wedeco)	Industry, reservoir replenishment
14		Ulu Pandan	2007	170,000	Secondary effluent	0.3 mm pre-screen (Amiad)	Pall (Microza UNA-620A)	Hydranautics (ESPA2)	UV (Wedeco)	Industry, reservoir replenishment
15		Changi	2010	228,000	Secondary effluent	Chloramination, auto-strainer	Siemens-Memcor (CP)	Toray	UV NaOH NaOCl	Industry, reservoir replenishment
16	Spain	El Prat de Llobragat	2007	15,000	Tertiary UV disinfected effluent	NaOCl	Zenon (Zeeweed 1000)	DOW (BW30-400 FR)	UV (Trojan)	Barrier against seawater intrusion
17		TIAS WWTP, Gran Canaria		1,000	Effluent	N/A	Siemens-Memcor	Filmtech	N/A	Irrigation
18	United Kingdom	Flag Fen	2000	1,600	Secondary effluent	0.15 drum screens	Pall (Microzoa USV6203)	Koch	None	Industry
19	USA	Water Factory 21, CA	1975-2004	19,000	Secondary effluent	Lime clarification Coagulation Flocculation Recarbonation Multimedia filtration	None	Koch	UV (Trojan)	Groundwater replenishment
20		West basin, El segundo, CA	1995	74,000	Secondary effluent	1 train: High rate clarification, mono media rapid filtration, Disinfection 3 Trains: None	Siemens-Memcor (CMF-S)	Trisep and Koch	UV (Trojan)	Industry, irrigation, groundwater recharge

	Country	Plant	Operation year	Capacity ($m^3.d^{-1}$)	Feed water	Pre-treatments	MF/UF	RO	Post-treatments	Applications
21		West Basin, Torrance, CA	1997	12,100	Secondary effluent	N/A	N/A	N/A	N/A	Industry
22		Livermore Water recycling facility	1997	2,800	Tertiary effluent	N/A	Siemens-Memcor	N/A	N/A	N/A
23		Scottsdale, water Campus, AZ	1999	46,000	Tertiary effluent	0.5 mm pre-screen	Siemens-Memcor (CMF-S)	Koch (8832 HR TFC Magnum)	Decarbonation Lime addition	Aquifer recharge, irrigation
24		West Basin, Carson	2000	19,000	Secondary effluent	N/A	Siemens-Memcor (CMF-S)	Koch and Hydranautics	N/A	Industry
25		Honouliuli WWTP	2000	8,000	Secondary effluent	N/A	Siemens-Memcor	N/A	None	N/A
26		Terminal Island, CA	2001	19,000	Secondary and tertiary effluent	Chloramination	Siemens-Memcor	Hydranautics	N/A	Industry
27		Orange County, CA	2004-2007	19,000	Secondary effluent	N/A	Siemens-Memcor (CMF-S)	Hydranautics ESPA2	UV (Trojan)	Groundwater replenishment
28		Alamitos Barrier WRD	2005	3,500	Tertiary effluent	0.5 mm strainers	Pall ((Microza UNA-620A)	Hydranautics	UV (Trojan) NaOCl	Protection of sea intrusion to groundwater
29		GWR Orange County, CA	2007	280,000	Secondary effluent	2 mm pre-screen	Siemens-Memcor (CMF-S)	Hydranautics ESPA2	AOP:UV (Trojan) +H_2O_2	Groundwater replenishment

APPENDIX 2: O&M PARAMETERS FOR THE NINE SURVEYED PLANTS

Plant name	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F	Plant G	Plant H	Plant I
	Generality								
Country	UK	Australia	USA	Singapore	Singapore	Spain	Kuwait	Belgium	USA
Reuse application	Idustry	Industrial application and reservoir replenishment	Groundwater recharge for Indirect potable reuse	Industry	Industry	Seawater intrusion barrier	Irrigation	Aquifer recharge for indirect potable reuse	Groundwater replenishment and seawater intrusion barrier
Start up year	2000	2008	2005	2007	2010	2007	2005	2002	2008
Design capacity (MLD)	1.6	66	11.4	150	232	15	375	7	329
Wastewater Type	Secondary effluent with nitrification	Secondary effluent with N removal	Secondary effluent	Secondary effluent	Secondary effluent	Secondary effluent N&P removal	Secondary effluent with N&P removal	Secondary effluent with N&P removal	Secondary effluent
	Pre-treatments								
Pre-filtration	Yes	N		Y	Y	Yes	Y	Y	Yes
Type/model	Hydrotech HDF 803 drum screen		Strainers SP Kinney AFW-1	Amiad ABF 10,000 Brush filter	Amiad ABF15,000	Disc-filter	Hydrotech disc	Longitudinal	Rotating gravity screen
Mesh size	0.15 mm		0.5 mm	0.5 mm		10 um	0.06 mm	1 mm	2 mm
Phosphorus removal	No	Yes	No			Yes	N	N	No
Type		Coagulation/Flocculation/lamella clarifier				Coagulation pre-disc filter			
Chemical used		FeCl3				PAX-18			
Dose (mg/L)						7 mg/L			
Chloramination/Chlorination/Disinfection	No	Y	No			Yes	N	Yes	Yes
Type		Chloramination		Chloramination	Chloramination	NaOCl		Chlorination pre -screen + addition of ammonia pre-MF	Chloramine
Dose (mg/L)						na		3.6	5

Plant name	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F	Plant G	Plant H	Plant I
Other pre-treatment		N				UV	Removal of dissolved organic substance and generation of a loose fiber cake on the membrane surface, Addition of coagulant: Ferric sulphate (2 ppm as Fe), dosed between the drumfilters and the UF	N	No
	Microfiltration/Ultrafiltration								
Membrane technology	Asai Kasei/Pall	Asahi Kasei/Pall	Asahi Kasei/Pall	Asahi Kasei/Pall	Siemens/Memcor	GE/ZENON	Norit/X Flow	GE/Zenon	Siemens/Memcor
Membrane model	Microza USV6203	UNA-620 A		Microza	CP	Zeeweed 1000	XIGA	Zeeweed 500c	CMF-S
Pore size	0.1	0.2	0.1	0.1	0.04	0.02	0.03	0.02	0.04
Membrane configuration *Immersed/pressurised*	Pressurised	Pressurised	Pressurised	Pressurised	Pressurised	Immersed	Pressurised	Immersed	Immersed
Total membrane area (m2)	1700	85400	3200	160000	360480	5016	304640	15600	730000
No. modules per unit/stack	17		8			60	4	26	608
No. stacks/units per train/tank	2		25			1	32	6	10
No. trains/tanks			1			2	68	5	4
Flux (LMH)									
Mean	35	65	60	44	47	27.8	65	25	33
Minimum	28						65	20	
Maximum	44						72	34	
Transmembrane pressure range Backwash	0.1	0.3	0.3-2	0.24	0.8	-0.1 to -0.55 bar	0.2-0.8	0.3-0.5	0.25 (0.21-0.9)

Plant name	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F	Plant G	Plant H	Plant I
Backwash frequency (min)	12	20	20-30	30	30	38	25	8-10	22
Backwash duration (s)	90		120			210	40	30	180
Backwash type	Water/Air	Water/Air	Water/Air			Water/Air	Water	Water	Water/air
Backwash water flux (LMH)	118					0.86	250		76.4
Backwash air flux (LMH)	na					18			275
Relaxation duration						na	0		0
Recovery of the process (%)	91		95			89.5	90	87	88-90
CEB									
Frequency (per month)	2	1 per day/1 per week					1 per day for both	Every 30-35 BW	Na
Duration (total downtime)(hours)	1.5						10 min each	30 s	
Chemical type	NaOCl	NaOCl/Citric acid					Cl2 + NaOH/H2SO4	Hypo	
Concentration (mg/L)							200@ pH 11/pH 2	200	
Temperature (oC)							ambient for both	ambient	
CIP		No							
Chemical type	NaOCl+NaOH/ Acid		Citric / NaOH			NaOCl/Citric acid	oxalic acid +ascorbic acid	NaOCl/ citric acid	Caustic soda+ Memclean/Citri c acid
Frequency (per month)	1-2		both 6 per year			1/2	1 per year	1 per month	Every 21 days
Duration (total downtime)(hours)			3d/3d			6h/6h	3-4 hours	4 hours	4 hours each
Concentration (mg/L)						500/1000	0.5% + 0.2%	200/na	2% + 0.5%/ 2%
Temperature (oC)						na/na	ambient		38
Membrane age									
Warrantied						3	7		7
Actual	4		7			3	5	7	
	Reverse osmosis								
Membrane technology	Koch	Toray	Hydranautics	Hydranautics	Toray	DOW	Toray	Dow	Hydranautics
Membrane model	TFC ULP	TML-20	ESPA2	ESPA2		BW30-400 FR	TML-20	BW 30LE-440	ESPA2

Plant name	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F	Plant G	Plant H	Plant I
Membrane configuration									
Total membrane area (m2)	1451	137500	43200	371280		2433		4002	580000
No. element per vessel	6	7	7	7		6		6	7
No. stages	2	3	2	2	2	2		2	3
No. vessel per stage						na			
Stage 1	4	120	72	64		na		21	78
Stage 2	2	60	36	36		na		11	48
Stage 3, if applicable		30				na			24
No. of trains	2	4	1	13	10	na	42	2	15
Flux (LMH)									
Mean	25	20	16.8	18	17	21		20	20
Minimum	21								
Maximum	30								
Recovery of the process (%)									
Mean	80	85	84	80	75	75	85		85
Minimum	75		80					75	80
Maximum	85		85					80	85.5
Feed pressure (bar)				6.5					
Stage 1	9-15	10	7.58-17.2			15		10-12	10.3-13.8
Stage 2	9-11								varies
Stage 3, if applicable									varies
Differential pressure (bar)									
Stage 1	1.2					na			2.4
Stage 2	0.8								1.2
Stage 3, if applicable									1.5
pH correction	Yes	No	No	Yes	Yes	No		Yes	Yes
pH achieved	6.8			6.8				7.25	6.8
Acid used	Sulphuric 77%							Sulphuric acid	Sulphuric
Concentration (mg/L)								40	

Plant name	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F	Plant G	Plant H	Plant I
Sodium bisulphate	No	N			Yes	Yes		Yes	No
Dose (mg/L)						na		if needed	
Antiscalant	Yes	Yes	Yes	na	Yes	Yes		Yes	Yes
Chemical used	Accepta 2651		Pretreat Plus 100			na			AWC A 102 Plus
Dose (mg/L)	22.5 to 1		3			5L/h		2.4	3.6
CIP									
Chemical type	Accepta 2068/Accepta 2066 +Accepta 2067		Citric /high pH dtergent			na		NaOH/Citric acid or biocide(DNBPA)	STTP/DDBS
Frequency (per month)	every 6 months for both		both 6 per year			na		4-6 per year for both	Every 6 months
Duration (total downtime)(hours)	2h/2h		3/3			na		0.5%/0.8%	8h/8h
Concentration (mg/L)	4%/3%					na		35 C/25 C	3%, pH 12/ 0.3%
Temperature (oC)	30 C/30 C		40			na			35 C/35C
Membrane age									
Warrantied	na					3			3
Actual	2		7			3		6	2
	Post treatment								
UV/AOP		Y			Yes		N	N	
Technology		UV/H2O2	UV		UV	UV			UV/H2O2
Model		Trojan UVPhox	Trojan UVPhox			Trojan UV			Trojan UVPhox
UV dose, mJ/cm2		500	500-1000			50-70			>300
Chemical dose (mg/L)		H2O2	no			None			3
pH correction		N	Yes		Yes		Y	Y	
Degassing tower			Yes		No	N		N	Y
Chemical used			No		NaOH	N	Caustic soda	NaOH	Lime and small amount of anionic polymer
Dose, mg/L								13	1
pH achieved			7.5					7.5	9
Rehardening		Calcium	No		No		N	N	

Plant name	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F	Plant G	Plant H	Plant I
Used chemical		bicarbonate				N			N
Dose, mg/L									
Disinfection			Yes			No	Y	N	No
Used chemical		NaOCl	Sodium hypochlorite		NaOCl				
Dose, mg/L			1						
Other post-treatment						Blend 50/50 with UF water		N	N
	Energy demand (kWh/m^3)								
Pre-treatment						0.1	<0.01		0.0012
Microfiltration/Ultrafiltration							0.05	0.177	0.26
Reverse osmosis						1.2		0.628	0.52
Post treatment						na			0.084
Total	1-1.14					1.3			1.06
	Mean OPEX								
Pre-treatment									
Microfiltration/Ultrafiltration									
Reverse osmosis									
Post treatment									
Total						0.36 euros/m^3		0.4 euro/m^3	

Plant name	Plant F	Plant A	Plant I	Plant C	Plant B	Plant G	Plant H	Plant E	Plant D
	Generality								
Country	Spain	UK	USA	USA	Australia	Kuwait	Belgium	Singapore	Singapore
Reuse application	Seawater intrusion barrier	Idustry	Groundwater replenishment and seawater intrusion barrier	Groundwater recharge for Indirect potable reuse	Industrial application and reservoir replenishment	Irrigation	Aquifer recharge for indirect potable reuse	Industry	Industry
Start up year	2007	2000	2008	2005	2008	2005	2002	2010	2007
Design capacity (MLD)	15	1.6	329	11.4	66	375	7	232	150

Plant name	**Plant A**	**Plant B**	**Plant C**	**Plant D**	**Plant E**	**Plant F**	**Plant G**	**Plant H**	**Plant I**
Wastewater Type	Secondary effluent N&P removal	Secondary effluent with nitrification	Secondary effluent	Secondary effluent	Secondary effluent with N removal	Secondary effluent with N&P removal	Secondary effluent with N&P removal	Secondary effluent	Secondary effluent
Pre-treatments									
Pre-filtration	Yes	Yes	Yes		N	Y	Y	Y	Y
Type/model	Disc-filter	Hydrotech HDF 803 drum screen	Rotating gravity screen	Strainers SP Kinney AFW-1		Hydrotech disc	Longitudinal	Amiad ABF15,000	Amiad ABF 10,000 Brush filter
Mesh size	10 um	0.15 mm	2 mm	0.5 mm		0.06 mm	1 mm		0.5 mm
Phosphorus removal	Yes	No	No	No	Yes	N	N		
Type	Coagulation pre-disc filter				Coagulation/Flocculation/lamella clarifier				
Chemical used	PAX-18				FeCl3				
Dose (mg/L)	7 mg/L								
Chloramination/Chlorination/Disinfection	Yes	No	Yes	No	Y	N	Yes		
Type	NaOCl		Chloramine		Chloramination		Chlorination pre -screen + addition of ammonia pre-MF	Chloramination	Chloramination
Dose (mg/L)	na		5				3.6		
Other pre-treatment	UV		No		N	Removal of dissolved organic substance and generation of a loose fiber cake on the membrane surface, Addition of coagulant: Ferric sulphate (2 ppm as Fe), dosed between the drumfilters and the UF	N		

Plant name	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F	Plant G	Plant H	Plant I
Microfiltration/Ultrafiltration									
Membrane technology	GE/ZENON	Asai Kasei/Pall	Siemens/Memcor	Asahi Kasei/Pall	Asahi Kasei/Pall	Norit/X Flow	GE/Zenon	Siemens/Memcor	Asahi Kasei/Pall
Membrane model	Zeeweed 1000	Microza USV6203	CMF-S		UNA-620 A	XIGA	Zeeweed 500c	CP	Microza
Pore size	0.02	0.1	0.04	0.1	0.2	0.03	0.02	0.04	0.1
Membrane configuration *Immersed/pressurised*	Immersed	Pressurised	Immersed	Pressurised	Pressurised	Pressurised	Immersed	Pressurised	Pressurised
Total membrane area (m2)	5016	1700	730000	3200	85400	304640	15600	360480	160000
No. modules per unit/stack	60	17	608	8		4	26		
No. stacks/units per train/tank	1	2	10	25		32	6		
No. trains/tanks	2		4	1		68	5		
Flux (LMH)									
Mean	27.8	35	33	60	65	65	25	47	44
Minimum		28				65	20		
Maximum		44				72	34		
Transmembrane pressure range	-0.1 to -0.55 bar	0.1	0.25 (0.21-0.9)	0.3-2	0.3	0.2-0.8	0.3-0.5	0.8	0.24
Backwash									
Backwash frequency (min)	38	12	22	20-30	20	25	8-10	30	30
Backwash duration (s)	210	90	180	120		40	30		
Backwash type	Water/Air	Water/Air	Water/air	Water/Air	Water/Air	Water	Water		
Backwash water flux (LMH)	0.86	118	76.4			250			
Backwash air flux (LMH)	18	na	275						
Relaxation duration	na		0			0			
Recovery of the process (%)	89.5	91	88-90	95		90	87		
CEB									
Frequency (per month)		2	Na		1 per day/1 per week	1 per day for both	Every 30-35 BW		

Plant name	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F	Plant G	Plant H	Plant I
Duration (total downtime)(hours)		1.5				10 min each	30 s		
Chemical type		NaOCl			NaOCl/Citric acid	Cl2 + NaOH/H2SO4	Hypo		
Concentration (mg/L)						200@ pH 11/pH 2	200		
Temperature (oC)						ambient for both	ambient		
CIP					No				
Chemical type	NaOCl/Citric acid	NaOCl+NaOH/ Acid	Caustic soda+ Memclean/Citric acid	Citric / NaOH		oxalic acid +ascorbic acid	NaOCl/ citric acid		
Frequency (per month)	1/2	1-2	Every 21 days	both 6 per year		1 per year	1 per month		
Duration (total downtime)(hours)	6h/6h		4 hours each	3d/3d		3-4 hours	4 hours		
Concentration (mg/L)	500/1000		2% + 0.5%/ 2%			0.5% + 0.2%	200/na		
Temperature (oC)	na/na		38			ambient			
Membrane age									
Warrantied	3		7			7			
Actual	3	4		7		5	7		
Reverse osmosis									
Membrane technology	DOW	Koch	Hydranautics	Hydranautics	Toray	Toray	Dow	Toray	Hydranautics
Membrane model	BW30-400 FR	TFC ULP	ESPA2	ESPA2	TML-20	TML-20	BW 30LE-440		ESPA2
Membrane configuration									
Total membrane area (m2)	2433	1451	580000	43200	137500		4002		371280
No. element per vessel	6	6	7	7	7		6		7
No. stages	2	2	3	2	3		2	2	2
No. vessel per stage	na								
Stage 1	na	4	78	72	120		21		64
Stage 2	na	2	48	36	60		11		36
Stage 3, if applicable	na		24		30				
No. of trains	na	2	15	1	4	42	2	10	13
Flux (LMH)									

Plant name	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F	Plant G	Plant H	Plant I
Mean	21	25	20	16.8	20		20	17	18
Minimum		21							
Maximum		30							
Recovery of the process (%)									
Mean	75	80	85	84	85	85		75	80
Minimum		75	80	80			75		
Maximum		85	85.5	85			80		
Feed pressure (bar)									6.5
Stage 1	15	9-15	10.3-13.8	7.58-17.2	10		10-12		
Stage 2		9-11	varies						
Stage 3, if applicable			varies						
Differential pressure (bar)									
Stage 1	na	1.2	2.4						
Stage 2		0.8	1.2						
Stage 3, if applicable			1.5						
pH correction	No	Yes	Yes	No	No		Yes	Yes	Yes
pH achieved		6.8	6.8				7.25		6.8
Acid used		Sulphuric 77%	Sulphuric				Sulphuric acid		
Concentration (mg/L)							40		
Sodium bisulphate	Yes	No	No		N		Yes	Yes	
Dose (mg/L)	na						if needed		
Antiscalant	Yes	Yes	Yes	Yes	Yes		Yes	Yes	na
Chemical used	na	Accepta 2651	AWC A 102 Plus	Pretreat Plus 100					
Dose (mg/L)	5L/h	22.5 to 1	3.6	3			2.4		
CIP									
Chemical type	na	Accepta 2068/Accepta 2066 +Accepta 2067	STTP/DDBS	Citric /high pH dtergent			NaOH/Citric acid or biocide(DNBPA)		
Frequency (per month)	na	every 6 months for both	Every 6 months	both 6 per year			4-6 per year for both		

Plant name	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F	Plant G	Plant H	Plant I
Duration (total downtime)(hours)	na	2h/2h	8h/8h	3/3			0.5%/0.8%		
Concentration (mg/L)	na	4%/3%	3%, pH 12/ 0.3%				35 C/25 C		
Temperature (oC)	na	30 C/30 C	35 C/35C	40					
Membrane age									
Warrantied	3	na	3						
Actual	3	2	2	7			6		
Post treatment									
UV/AOP					Y	N	N	Yes	
Technology	UV		UV/H2O2	UV	UV/H2O2			UV	
Model	Trojan UV		Trojan UVPhox	Trojan UVPhox	Trojan UVPhox				
UV dose, mJ/cm2	50-70		>300	500-1000	500				
Chemical dose (mg/L)	None		3	no	H2O2				
pH correction				Yes	N	Y	Y	Yes	
Degassing tower	N		Y	Yes			N	No	
Chemical used	N		Lime and small amount of anionic polymer	No		Caustic soda	NaOH	NaOH	
Dose, mg/L			1				13		
pH achieved			9	7.5			7.5		
Rehardening				No	Calcium bicarbonate	N	N	No	
Used chemical	N		N						
Dose, mg/L									
Disinfection	No		No	Yes		Y	N		
Used chemical				Sodium hypochlorite	NaOCl			NaOCl	
Dose, mg/L				1					
Other post-treatment	Blend 50/50 with UF water		N				N		
Energy demand (kWh/m^3)									
Pre-treatment	0.1		0.0012			<0.01			
Microfiltration/Ultrafiltra			0.26			0.05	0.177		

Plant name	Plant A	Plant B	Plant C	Plant D	Plant E	Plant F	Plant G	Plant H	Plant I
tion									
Reverse osmosis	1.2		0.52				0.628		
Post treatment	na		0.084						
Total	1.3	1-1.14	1.06						
Mean OPEX									
Pre-treatment									
Microfiltration/Ultrafiltration									
Reverse osmosis									
Post treatment									
Total	0.36 euros/m^3						0.4 euro/m^3		