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BOTTLENECKS OF SUSTAINABLE WATER REUSE: ADVANCED TREATMENT OF MUNICIPAL WASTEWATER AND MEMBRANE CONCENTRATES

KNELPUNTEN BIJ DUURZAAM HERGEBRUIK VAN WATER: DOORGEDREVEN ZUIVERING VAN HUISHOUDELIJK AFVALWATER EN MEMBRAANCONCENTRATEN

door

ir. Kris VAN HEGE

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op gezag van
Rector: **prof. dr. apr. A. DE LEENHEER**

Decaan:
prof. dr. ir. O. VAN CLEEMPUT

Promotor:
prof. dr. ir. W. VERSTRAETE

Ghent, May 2002

Promotor:	Author:
prof. dr. ir. Willy Verstraete	ir. Kris Van Hege

The research was conducted at the Laboratory Microbial Ecology and Technology (Ghent University), Coupure links 653, 9000 Ghent. Tel: +32 9 264 59 76, Fax: +32 9 264 62 48

WOORD VOORAF

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Kris.

NOTATION INDEX

AC	Activated Carbon
AHPA	American Public Health Association
AOP	Advanced Oxidation Process
AWWA	American Waterworks Association
BAC	Biological Activated Carbon
BDD	Boron-Doped Diamond
BioMAC	Biological Membrane Assisted Carbon filtration
BOD	Biochemical Oxygen Demand
CF	Concentration Factor
CFU	Colony Forming Units
COD	Chemical Oxygen Demand
D	Dalton
DBP	Disinfection By-Products
DM	Dry Matter
DO	Dissolved Oxygen
E. coli	*Escherichia coli*
EC	Electrical Conductivity
EDX	Energy Dispersive X-ray analysis
EEA	European Environmental Agency
EPA	Environmental Protection Agency
ETC/IW	European Topic Centre on Inland Waters
EU	European Union
FC	Faecal Coliforms
GAC	Granular Activated Carbon
GC-MS	Gas Chromatography - Mass Spectrometry
IAHR	International Association of Hydraulic Research
IPCC	Intergovernmental Panel on Climate Change
IWA	International Water Association
KMI	Koninklijk Meteorologisch Instituut
KVE	Kolonie-Vormende Eenheden

MBR	Membrane Bioreactor
MF	Microfiltration
NDMA	N-nitrosodimethylamine
NF	Nanofiltration
NTU	Nephelometric Turbidity Unit
PAC	Powdered Activated Carbon
RO	Reverse Osmosis
RWZI	Rioolwaterzuiveringsinstallatie
SAT	Soil Aquifer Treatment
SD	Standard Deviation
SEM	Scanning Electron Microscope
SHE	Standard Hydrogen Electrode
SOC	Synthetic Organic Chemical
SOP	Sensorial Odour Perception
SOP_{av}	Average Sensorial Odour Perception per litre gas
SS	Suspended Solids
TAN	Total Ammonia Nitrogen
TDS	Total Dissolved Solids
TMP	Trans Membrane Pressure
TOC	Total Organic Carbon
UASB	Upflow Anaerobic Sludge Blanket
UF	Ultrafiltration
UNEP	United Nations Environmental Program
US	United States
UV	Ultra Violet radiation
VOC	Volatile Organic Component
VLAREM	Vlaams Reglement betreffende de Milieuvergunning
WCED	World Commission on Environment and Development
WEF	Water Environment Federation
WHO	World Health Organisation
WWTP	Wastewater Treatment Plant

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INTRODUCTORY CHAPTER

BOTTLENECKS OF SUSTAINABLE WATER REUSE: ADVANCED TREATMENT OF MUNICIPAL WASTEWATER AND MEMBRANE CONCENTRATES

AIM OF THE STUDY

It was the aim of the present thesis to provide an insight into the various aspects of municipal wastewater reclamation and reuse, to review and evaluate existing advanced treatment technologies, and to explore and develop alternative approaches. Therefore, an overview of the existing technologies was made, and the major bottlenecks were identified. Subsequently, 3 different technological approaches for the reclamation of municipal wastewater were developed and evaluated:

1. As an alternative to filtrative desalination, evaporative treatment of WWTP effluent was considered for the direct production of high quality reclaimed wastewater.
2. Advanced effluent polishing was carried out using an innovative filtrative/adsorptive biological reactor.
3. Electrolysis was introduced as a novel end-of-pipe technology for the destruction of reverse osmosis concentrates.

OVERVIEW

In the first Chapter, an insight is given into the commodity “water”. The factors influencing the availability as well as the quality of water are briefly discussed. It is shown that the present state of the asset water displays strong indications of lack of sustainability. Because the traditional resources are fully exploited in many areas, new and alternative water resources have to be explored. In this respect, it has become apparent that the only “new” water resource is wastewater and therefore the concepts of wastewater reclamation, reuse and recycling are introduced in Chapter 2. It is demonstrated that wastewater reclamation and reuse can be incorporated in the natural water cycle. As a possible source, the effluent of municipal wastewater treatment plants is considered and a number of possible reuse applications are summarised. It is recognised that the quality of reclaimed water is of principal concern; therefore a number of microbiological, chemical and organoleptic parameters are evaluated with respect to possible adverse effects. Finally, some existing wastewater reclamation guidelines are reviewed. Chapter 3 gives an overview of the available treatment techniques. Primary and secondary treatment as well as disinfection technologies are shortly discussed. Because of its importance with respect to the production of high-quality reclaimed water, the role of tertiary (or advanced) treatment is surveyed more elaborately. More specifically, membrane processes, evaporation and electrochemical treatment are analysed as possible wastewater reclamation technologies, and some bottlenecks are identified. Membrane processes have been extensively employed for the reclamation of municipal wastewater, but membrane fouling and the treatment of the produced concentrate remain major problems. Evaporation allows extensive decontamination of WWTP effluent, but care has to be taken with respect to volatile components. Finally, electrolysis allows the incineration of organic pollutants yet can yield organochlorinated constituents. In Chapter 4, evaporation is proposed as an alternative wastewater reclamation technology for the direct production of high-quality reclaimed water. In order to eliminate the volatile components present in WWTP effluent, Fenton oxidation was evaluated as a pre-treatment. Chapter 5 encompasses the implementation of an innovative combined biological activated carbon and microfiltration reactor for advanced effluent polishing treatment. The reactor was tested with regard to the removal of organic and biological compounds and its capacity to attenuate fluctuations in composition of the feed solution. In Chapter 6, the end-of-pipe treatment of reverse osmosis concentrates is envisaged. In addition to biological treatment, the ability of electrochemical elimination of the organic and inorganic constituents is investigated. Chapter 7 presents an overall evaluation of the tested technological solutions. Finally, some recommendations with respect to the emergence of new pollutants and advanced technological treatment solutions are formulated.

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CHAPTER 1

WATER, THE BLUE GOLD

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1. INTRODUCTION

Clean fresh water is the most precious commodity on earth - life simply cannot exist without it. Water plays a key role in the aquatic, wetland and terrestrial components of the global ecosystem. Individuals and communities need water for a wide range of uses:

- For basic needs (for drinking, cooking, washing, 5 L of water per capita per day is considered absolutely indispensable).
- For reasonable quality of life and good community health (toilet flushing, waste disposal, washing requires around 80 L per capita per day).
- To generate and sustain wealth (commercial fishing, aquaculture, agriculture, power generation, industry, transport and tourism).
- For recreation (sport fishing, boating, swimming).

Despite the fact that water is the most renewable of our essential resources (as exemplified by the hydrologic cycle), it has become clear that an adequate supply of unpolluted water can in many cases no longer be taken for granted (Birkett, 2000). In many of the world's regions, the impact of humanity on the natural water cycle is causing an imbalance in the ecosystem upon which its very own existence depends. Therefore, it is becoming vital to carefully look after the available water supplies, to use them frugally and to prevent their contamination. Where the traditional resources are being fully exploited, new and alternative ways of water conservation have to be explored (Asano, 1994).

It is the aim of the present chapter to give an insight into the availability and the use of the resource "water". The impact of human activities on water quality and quantity is discussed, specifically focusing on the current European situation. Finally, the concept of sustainable water management is described and new tools for managing this vital asset, *i.e.* wastewater reclamation and reuse, are introduced.

2. STATE OF THE WATER RESOURCES

2.1. Water availability

The formation of water on our planet is thought to have occurred during the Precambrian period, 3.4 billion years ago. The total quantity of water is now nearly constant at $1.386*10^9$ km^3. However, the distribution of the Earth's water shows that far less than 1 % is potentially directly available as fresh water (Table 1).

Table 1 Distribution of the Earth's water (Shiklomanov, 1993)

	Percentage	**Volume (10^6 km^3)**
Saltwater stock (in oceans, groundwater, lakes)	97.48	1351
Freshwater stock, of which:	2.52	35.03
Snow and ice	*69.5*	*24.36*
Fresh groundwater	*30.1*	*10.53*
Rivers, wetlands and freshwater lakes	*0.3*	*0.11*
Miscellaneous	*0.1*	*0.03*

To assess the amount of water available per capita, the "total renewable freshwater resource" is calculated as the endogenous resource (*i.e.* the run-off generated internally *e.g.* by precipitation) plus the exogenous resource (*i.e.* the water imported from upstream countries) (EEA, 1999b). Based on this definition, the world average is 7300 m^3/(capita*year), while the average internal water resource is estimated to be 3200 m^3/(capita*year) for the European Union (EEA, 2000). The relative amount of water resources available per capita is often divided into a number of categories as shown in Table 2.

Table 2 Classification of relative per capita water availability (EEA, 1999b)

Category	**Water availability (m^3 per capita per year)**
Extremely low	< 1000
Very low	1 000 - 2 000
Low	2 000 - 5 000
Medium	5 000 - 10 000
Above medium	10 000 - 20 000
High	20 000 - 50 000
Very high	> 50 000

Unfortunately, these resources are far from evenly distributed as rainfall and run-off are apportioned in both space and time in a grossly irregular manner. This is illustrated in Figure 1 for 23 European countries. According to Table 2, about half of the countries described in Figure 1 can be described as having a low per capita water availability. Freshwater accessibility is even worse in the Czech republic, Poland and Belgium, which fall into the very low availability category. Hydrologists designate countries, which fall into the latter category as "water-scarce" (Liessens *et al.*, 1996).

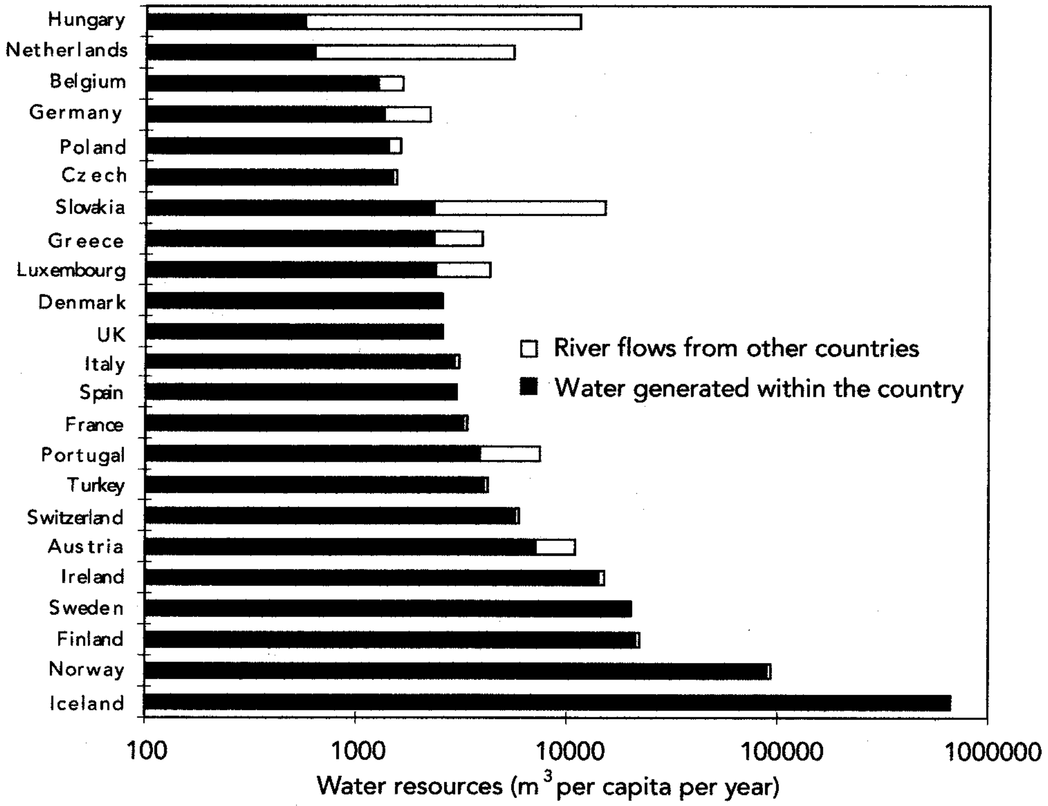

Figure 1 Freshwater availability in Europe (EEA, 2000)

2.2. Water demand

Global population projections suggest that the world population of 6.055 billion in 2000 will increase 20 % to 7.015 billion in 2015, and to 7.825 billion in 2025, a 30 % increase. In Asia and Africa, the urban population will double during this period (Okun, 2001). This, coupled to the economic development and increased irrigation, will result in a considerable increase in the projected total water demand (Figure 3). Because of the slowdown in the rate of growth of the main driving forces for water demand and improvements in the efficiency of water use, only a very slight increase is projected for Europe (EEA, 1999a).

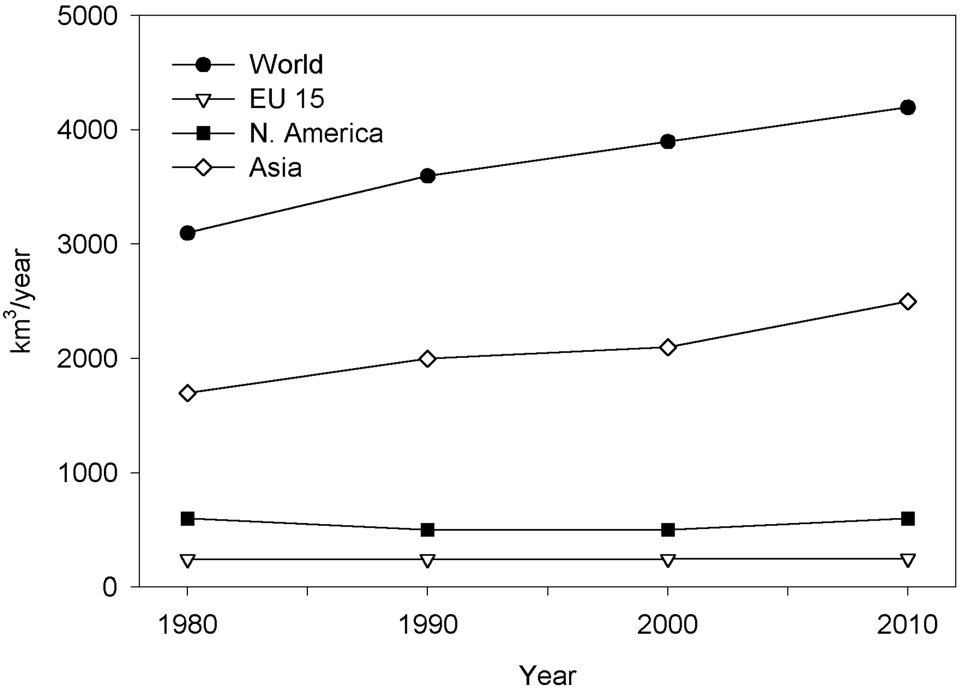

Figure 2 Total water demand - trends and projections (EEA, 1999a)

In Europe as a whole, only about one-fifth of the theoretically available water is abstracted (660 m³/(capita*year)). However, this assessment does not take into account the water needed to sustain aquatic life, nor does it account for the spatial dislocation of abstractions and returns. As depicted in Figure 3, in most European countries the amount of water available is far greater than the volume used, although some countries heavily rely on their renewable freshwater sources.

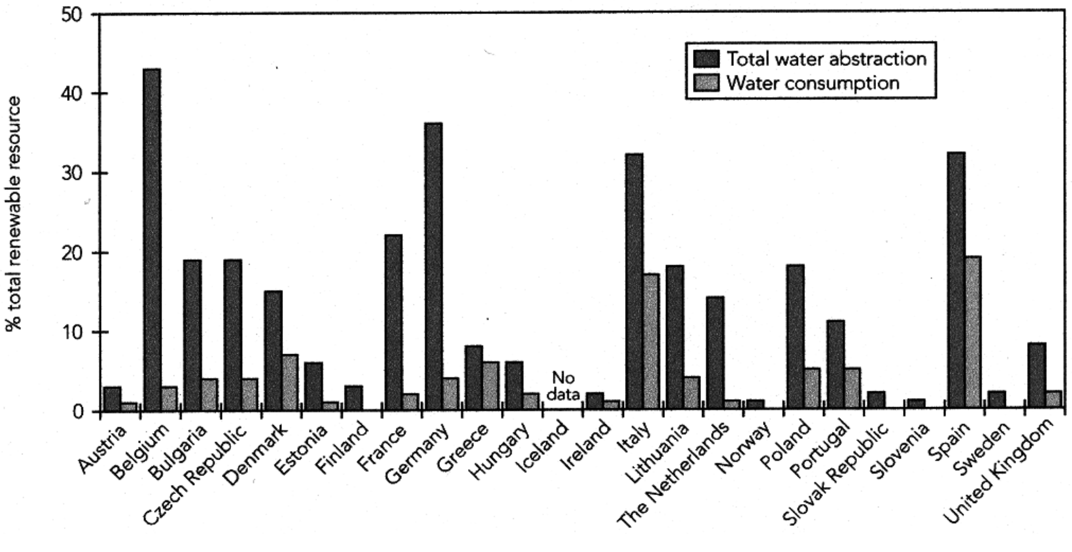

Figure 3 Intensity of water abstraction and water consumption as a percentage of the total renewable freshwater resources in Europe (EEA, 1999a)

According to international experts, water management has to be considered an important element in the national economy when the use of water exceeds 30 % of the water reserves (Lazarova *et al.*, 2000; Thoeye *et al.*, 2000; Vandaele *et al.*, 2000). Germany, Spain, Italy and Belgium fall into this category.

3. IMPACTS ON WATER RESOURCES

Human activities have a profound influence on the natural hydrologic cycle in 3 main ways (EEA, 2000; Thoeye *et al.*, 2000):

1. Abstraction and consumption of both surface and groundwater for agricultural, industrial and potable purposes.
2. Alterations to the environment:
    - Land sealing by urbanisation.
    - Agricultural drainage and flood control.
    - Damming and modification of river flow patterns.
3. Pollution:
    - Point source pollution from households and industry (*i.e.* discharges from sewage treatment works and industrial processes).
    - Diffuse pollution from urban and agricultural run-off, and pollution from waste disposal.

The share of these elements in the overall impact is difficult to measure, but they can affect the water resource in both a quantitative and qualitative way.

3.1. Quantitative impact

3.1.1. Water shortage and water stress

Water stress is generally related to an overproportionate abstraction of water in relation to the resources available in a particular area. Although water is considered scare and a high pressure on the internal resources exists in some European countries (*e.g.* Belgium), potentially all European countries have sufficient resources to meet their national demand (Figure 3). However, the national statistics tend to mask problems that may be occurring locally. In the agricultural areas of southern Europe and the urban conurbations centres in the Northwestern Europe, demand exceeds the available water (EEA, 2000). As a consequence, restraints on industrial and other uses are already occurring in many parts of Europe. For example, river basin authorities are increasingly restricting the rights of industry to abstract water.

3.1.2. Aquifer over-extraction

Two-thirds of the European citizens rely on ground water for their supplies of drinking water and other water needs. As a consequence, aquifer over-abstraction, resulting in a lowering of the groundwater table, is a considerable problem in many European countries and it is estimated that 140 million Europeans live in or near a region of such groundwater over-exploitation (EEA/UNEP, 1997). In the Western part of Flanders, this issue is eminently clear when studying the equilibrium heights of the groundwater tables Landeniaan and Sokkel (Thoeye *et al.*, 2000). The excessive water abstraction from these aquifers has resulted in a sharp decrease of the groundwater height as shown in Table 3.

Table 3 Decrease in groundwater equilibrium height for the Landeniaan and the Sokkel during the period 1965-1985 in a number of Flemish cities (De Ceukelaire *et al.*, 1992)

City	Landeniaan (m/10 year)	Sokkel (m/10 year)
Kortrijk	7	-
Roeselare	18	3.5
Ronse	3.5	3.5
Waregem	18	35
Aalst	-	65
Oudenaarde	-	15

In coastal aquifers, salt water intrusion already occurs in 9 European countries (ETC/IW, 1998) and also along the Flemish coast the groundwater extraction has reached its maximum capacity (Van Houtte *et al.*, 1998). Aquifer over-utilisation also gives rise to drying up of rivers and desiccation of wetlands - already 50 % of the major wetlands in Europe have an “endangered status” (EEA, 1999b).

3.1.3. Droughts and desertification

Although droughts are a normal, recurrent factor of the Europe’s climate, it is generally recognised that the impact of a drought is intensified in areas with the greatest pressures on their water resources, especially in those regions with high irrigation demands. The most recent period of drought - between 1988 and 1992 - caused irrigation problems over a large part of Europe, ranging from Hungary to Spain, where agricultural production was heavily affected and uneven distribution of water triggered inter-regional political concerns (EEA, 1999b).

Extended or recurrent droughts can aggravate desertification, caused by human activities such as over-exploitation of water and soil, fires and change of land use. In Europe, the semi-arid Mediterranean countries are particularly vulnerable to desertification, having mountainous terrain with steep slopes, over-exploited water resources and rain-fall with substantial erosion capacity (EEA, 1997).

3.1.4. Climate change

The climate change resulting from anthropogenic emissions of CO_2 and other greenhouse gasses may add to the pressure on water resources over the next few decades. Although it is impossible to precisely forecast Europe's future climate, model-based predictions show that there will be a temperature rise of 1 to 3.5 °C, together with an increase in precipitation in Northern Europe and a decrease in Southern Europe (IPCC, 1996). This may lead to a reduction in renewable water resources in the latter area and even in areas where precipitation would increase, the availability of water could decrease due to stronger evaporation. Despite the inherent uncertainty of such prophecies, it is generally acknowledged that the greater the stress this system is under at present, the more sensitive it will be to climate change (EEA, 1999b).

3.2. Qualitative impact

The impacts of both diffuse and point source pollution from agriculture, industry and households are very diverse and complex, but the general picture is one of deterioration in the ecological quality of Europe's aquatic systems (EEA, 1997).

3.2.1. Surface water

Phospohorus and organic matter concentrations have decreased significantly in Europe's large rivers due to the general improvement in wastewater treatment, particularly of sewage (Figure 4.a). Nitrate concentrations have rapidly increased between 1980 and 1990 beacuse of intensification of agriculture and urban wastewater treatment, but recent data indicate that the levels have remained relatively stable in many rivers since then (EEA, 2000). Also, strong indications exist that surface water acidification has been sucessfully abated through the implementation of controls on the atmospheric emissions of suplhur and nitrogen oxides (EEA/UNEP, 1997). However, despite the general reduction in organic pollution and the concomitant improvement in oxygen conditions - which is also observed in Flanders (Thoeye *et al.*, 2000) - the state of many rivers remains poor. Also, very little data exist with respect to the fate or impact of many xenobiotic components released into the aquatic environment.

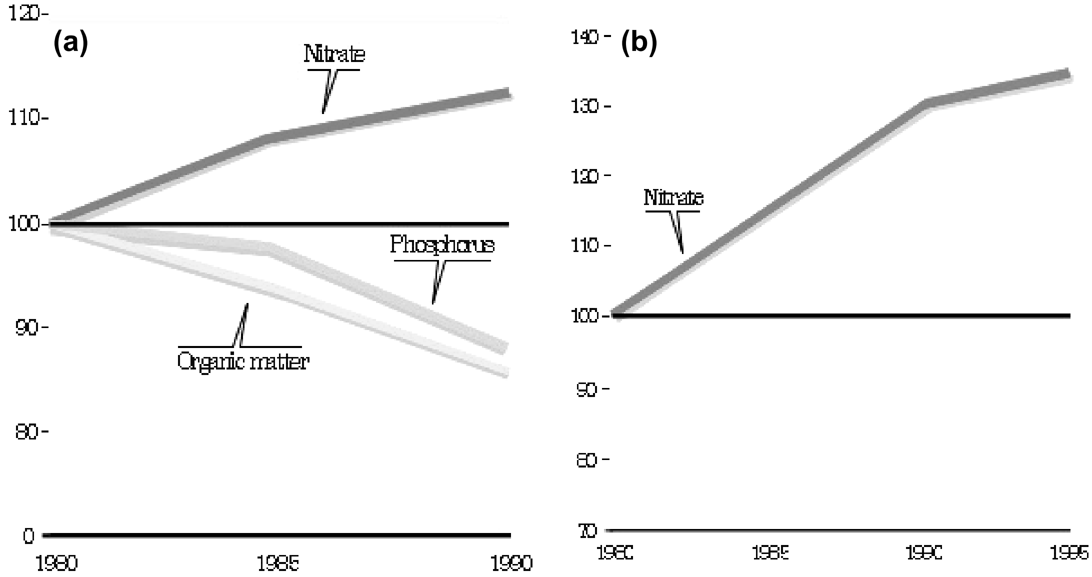

Figure 4 (a) Development of the organic matter, phosphorus and nitrate concentration in large European Union rivers and (b) the nitrate concentration in the groundwaters of France, UK and Denmark (EEA, 1995).

3.2.2. Groundwater

Notwithstanding the implementation of the European Nitrates directive, which sought to reduce or prevent pollution of water resulting from the application and storage of inorganic fertiliser and manure on farmland, groundwater nitrate levels still appear to be rising (Figure 4.b). In 1998, more than a quarter of the sampling sites exceeded the maximum admissible Drinking Water Directive of 50 mg NO_3^-/L in 13 % of Europe's regions (EEA, 1998). Apart from potential health effects, high levels of nitrates may also be a more important cause of groundwater acidification than atmospheric deposition, due to the occurrence of soil denitrification (EEA/UNEP, 1997; Thoeye *et al.*, 2000). Because of the long lag time between nitrogen use and its seeping into the groundwater stock, it is even likely that this situation will worsen before the reduced land usage begins to affect the concentrations in water. Pesticides are also causing difficulties as many different types have been detected in groundwater at levels greater than the Drinking Water Directive maximum allowable concentration of 0.1 µg/L. The commonly detected pesticides - atrazine, simazine and lindane - have also been detected in Flanders in relatively elevated concentrations (> 0.5 µg/L). Again, the lack of monitoring data, coupled to the relative persistence of such components has been reason for concern.

4. RESOURCE MANAGEMENT

From the previous it is clear that in many areas, conflicts arise between supply and demand or, in a wider framework, between human demand and ecological needs. No longer can mankind replenish its needs for water by merely augmenting the supply; balancing the supply and demand through integrated water management has become an imperative necessity.

4.1. Water management and sustainability

"Sustainable use" has been most memorably defined in the Brundtland report of the World Commision on Environment and Development (WCED, 1987) as "a use which meets the needs of the present without compromising the ability of future generations to meet their own needs" and is visualised in Figure 5. This concept has generally been interpreted in a way that economic development can no longer be separated from environmental issues and that human society should learn to live without eroding the stock of natural assets.

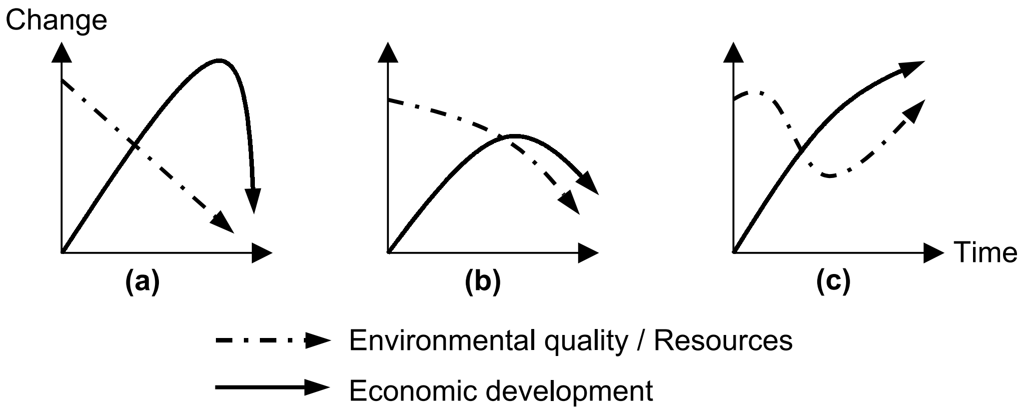

Figure 5 Concepts of (a) total collapse, (b) non-sustainable and (c) sustainable development (Beaufays and Dubois, 1999)

4.2. Tools for sustainable water management

As discussed before, water management - which covers the fields of water supply, urban hydrology and wastewater treatment - displays in its present form strong indications of lack of sustainability (Larsen and Gujer, 1997). Within the framework of sustainable water management it is therefore essential to provide tools for managing both the demand side as well as the supply side (Figure 6). Although conservation measures have effected reductions in the per capita demand, the increasing demand from population growth and industrial development dictates the necessity for more water (Okun, 2000). As in many areas the traditional supplies are fully exploited, new and alternative water resources are needed (Dewettinck *et al.*, 1999).

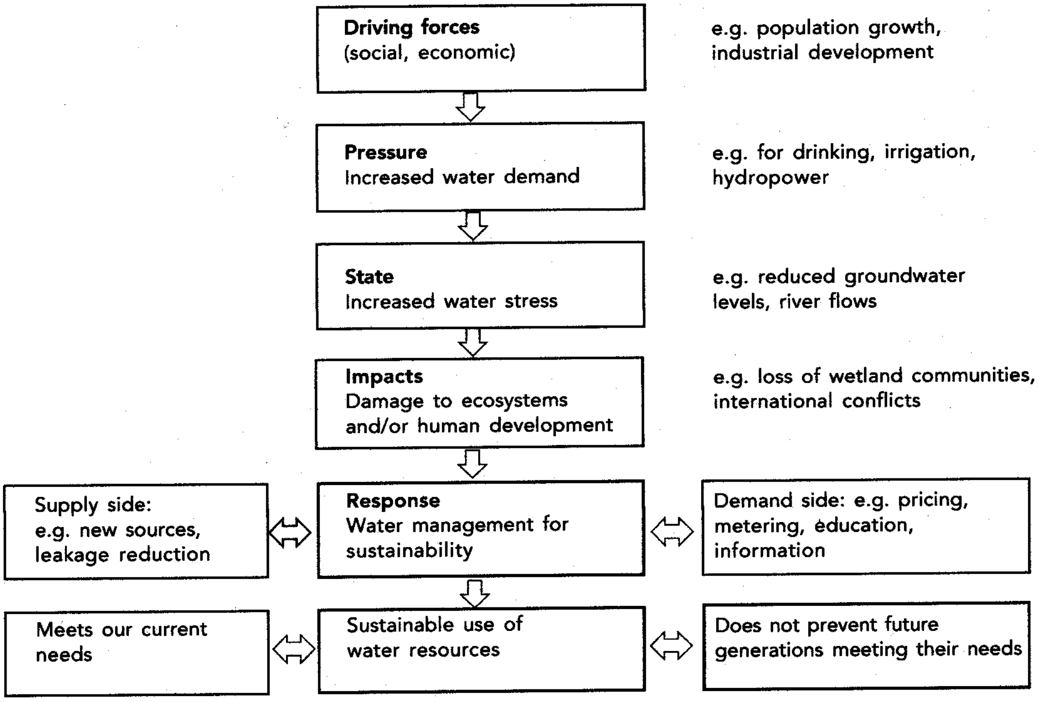

Figure 6 Water management for sustainability (EEA, 2000)

Especially in cities and other densely populated areas, it has become apparent that the only significant "new" resource is wastewater. As a consequence, water reclamation, reuse and recycling are receiving considerable attention as novel water resource management tools (Asano, 1994; Asano and Levine, 1995; Lazarova *et al.*, 2000; Okun, 2001). Reclaimed water serves in many regions in the world an important function in water supply management by providing a means to produce quality source water for irrigation, industrial and urban water requirements.

Within the afore-mentioned framework of sustainable development, the reuse and recycling of water and wastewater can even be considered self-evident. Indeed, all natural processes, such as the hydrologic cycle, are characterised by use and reuse, whereby the wastes of one step become the raw materials of the next. Therefore, an essential feature of sustainability is its cyclic character. As a consequence, the implementation of water reuse is of prime importance in the achievement of true sustainability in water resources management.

CHAPTER 2

WASTEWATER RECLAMATION, RECYCLING AND REUSE

1. DEFINITIONS (Asano, 1994; Asano and Levine, 1998)

Wastewater reclamation encompasses the treatment or processing of wastewater to make it reusable, and *water reuse* or *wastewater reuse* is the use of the treated wastewater for a beneficial application. The resulting product is designated *reclaimed wastewater*, although recently the less pejorative connotation *e-water* (economical / ecological water) was introduced (Roorda, 2001; Ruiters *et al.*, 1999). In contrast to reuse, *wastewater recycling* or *water recycling* normally involves only one user or use and the effluent from the user is captured and redirected back into that use scheme. In this context, the term water recycling is predominantly linked to industrial applications such as the pulp and paper production and the electricity generation.

The use of reclaimed wastewater where there is a direct link (*i.e.* a pipeline or another type of conveyance facility) between the treatment system and the reuse application is termed *direct reuse*. Hence, this term is associated with agricultural and landscape irrigation, industrial and urban applications and dual water systems. *Indirect reuse* involves dilution, dispersion, blending and attenuation of a reclaimed water by discharge into an impoundment, receiving water or groundwater aquifer prior to reuse such as in groundwater recharge or surface water replenishment. Indirect reuse can be planned or *incidental*. *Unplanned* indirect wastewater reuse, through disposal of treated effluent into a stream or groundwater basin and subsequent assimilation downstream, has been an accepted practice throughout the world for centuries. Indeed, in the densely populated areas of Europe, wastewater treatment plant effluent makes up a large portion of the normal flow of many rivers, which is then withdrawn for domestic and industrial water supplies. *Deliberate* or *planned* indirect reuse includes uses such as artificial groundwater recharge and river flow augmentation.

A differentiation is also made between potable and non-potable reuse applications: *potable water reuse* refers to the use of highly treated reclaimed water to augment drinking water supplies, whereas *non-potable reuse* comprises all other applications.

2. ROLE OF WASTEWATER RECLAMATION, RECYCLING AND REUSE IN THE WATER CYCLE

Supplying the anthropogenic water cycle with alternative water resources, such as reclaimed water, accelerates the natural water cycle through advanced technological solutions (Lazarova *et al.*, 2000). Figure 1 depicts the incorporation of the concepts of engineered treatment, wastewater reclamation and water reuse in the natural hydrologic cycle.

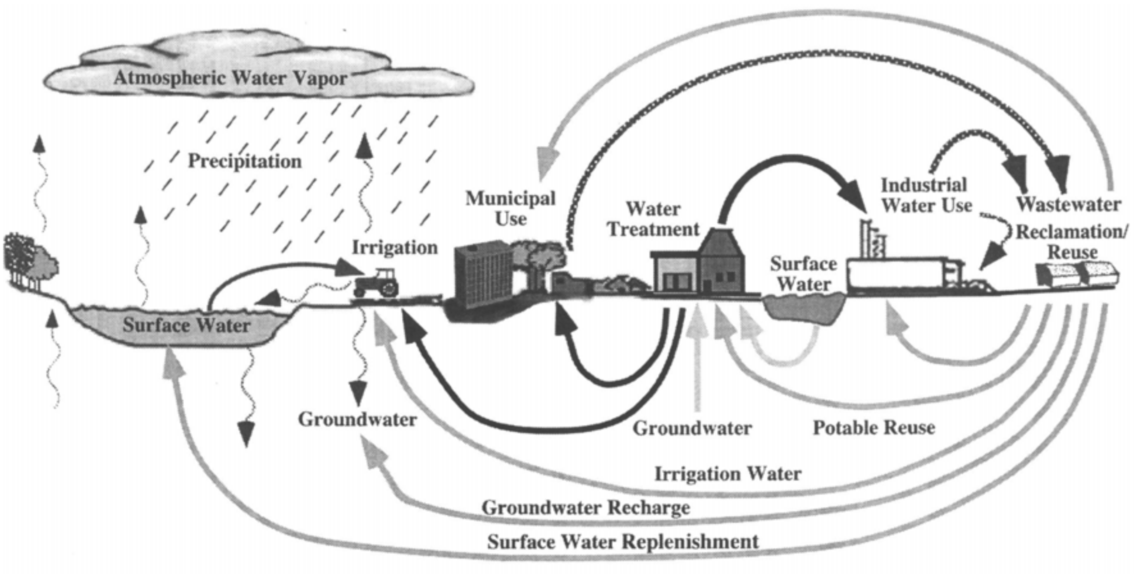

Figure 1 The role of engineered treatment, reclamation and water reuse facilities in the cycling of water through the hydrologic cycle (Asano and Levine, 1998)

In urban, industrial and agricultural areas, the concepts of wastewater reclamation, recycling and reuse result in the creation of closed and artificial loops in the hydrologic cycle. In this way, a dynamic balance between the human and ecological needs is ensured, while responding to both the present and the future demand. Thus, water reuse can be viewed as an integral element of sustainable water resource management.

3. SOURCES OF RECLAIMED WASTEWATER

In densely populated areas, one of the prime sources of reclaimed water is the effluent originating from centralised wastewater treatment plants. Several factors contribute to the suitability of this wastewater for various reuse applications:

- In the more developed countries, urban wastewater receives treatment prior to discharge. Under the United States Clean Water Act, treatment is intended to render the water "fishable and swimmable" (Okun, 2000) while in Europe, WWTP effluents must comply with the European Wastewater Directive standards in order to protect the aquatic environment (European Commission, 1991).

- As a result of treatment, in some cases the quality of the discharged effluent is higher than that of the receiving surface water body (Van Houtte *et al.*, 1998).
- Centralised WWTPs process tremendous amounts of water: Dutch WWTPs produced 1800*10^{6} m^{3} effluent in 1998 (Roorda, 2001), while in Flanders this amount totalled 523*10^{6} m³ in 2000 (Anonymous, 2000). With the exception of cooling water needs, this quantity easily surpasses the demand of any given water use in many countries. For example, in Flanders, the annual drinking water consumption is 439*10^{6} m³, while the industrial and agricultural groundwater extraction amounts to 285*10^{6} m³ per year (Thoeye *et al.*, 2000).
- The effluent is produced where the water demand is high, *i.e.* in cities and other densely populated areas. In addition, WWTPs are often situated in or near industrial parks.

4. WATER REUSE APPLICATIONS

In theory, wastewater can be reused for any purpose, only the level of treatment required to bring it to a quality appropriate for a certain need varies. The potential applications of reclaimed water can be divided in seven categories (Asano, 1994).

4.1. Agricultural irrigation

The largest single use of water world-wide is agricultural irrigation, which accounts for approximately 80 % of the consumptive water use (Gleick, 2000). Where water shortages exist, the use of reclaimed wastewater offers a reliable, valuable and low cost water resource. Additional benefits are: increased crop yields, decreased reliance on chemical fertilisers and increased protection against frost damage (Asano and Levine, 1995). Extensive studies conducted in California in the 1970s and 1980s, namely the Pomona Virus study and the Monterey Wastewater Reclamation Study for Agriculture conclusively demonstrated that virtually pathogen-free effluents could be obtained from municipal wastewater via tertiary treatment and chlorine disinfection (Bahman *et al.*, 1998). Hence, even food crops used for raw consumption could be irrigated with such a reclaimed municipal wastewater without any adverse environmental or health effect. Amahmid *et al.* (1999) found that cultures irrigated with effluent from sewage waste stabilisation ponds were free from contamination with *Giardia* cysts and *Ascaris* eggs.

The use of reclaimed wastewater for agricultural irrigation has become an essential water resource in arid regions, such as Saudi Arabia (Hussain and Al-Saati, 1999), Tunesia (Bahri, 1998), Israel (Oron, 1998), South Africa (Odendaal *et al.*, 1998) and Portugal (Marecos do Monte, 1998). California has substantial experience with the agricultural use of reclaimed water: in 2000, about one third of the more than 600 million m³ of reclaimed wastewater produced, was used for the irrigation of fodder, pasture, vineyards, row crops and organic vegetables (Gleick, 2000).

4.2. Landscape irrigation

The irrigation of landscaped areas in the urban environment, such as parks, schoolyards, freeway medians, cemeteries, golf courses and residential yards has become an important use of reclaimed water. Within this category, there has been a significant increase in the number of golf courses using reclaimed water (Asano, 1994).

Outdoor landscape irrigation is widely employed in California, where around 20 % of the reclaimed water is used for this application. For example, the reuse facility of the Irvine Ranch Water District can provide about 49000 m³ reclaimed water per day for diverse purposes including public and private landscape irrigation (Young *et al.*, 1998). In addition, the City and County of San Francisco is implementing a 40000 m³/day reclamation facility; of which two-thirds of the flow is targeted for landscape irrigation - the largest user being Golden Gate park (Jolis *et al.*, 1995; Jolis *et al.*, 1999).

4.3. Industrial applications

Some of the largest and highest-value wastewater uses are for industrial applications. Water recycling has been successfully implemented in a wide range of industrial settings, and in many other cases reclaimed wastewater has been used as an external water resource (Spencer, 2000). In most cases, reclamation and recycling processes can provide a higher and more consistent quality of water than is obtainable from many existing surface water supplies (Judd, 1999). The application of reclaimed wastewater is of particular interest in high volume industrial uses as cooling, boiler feed or process water. Examples include: power plants (Blackson and Moreland, 1998), pulp and paper mills (Chen and Horan, 1998; Mobius and Cordes-Tolle, 1994), electroplating and semiconductor industries (Shah and Ploeser, 1999), iron and steel production (Borsani *et al.*, 1996), textile processing (Ciardelli *et al.*, 2001; Li and Zhao, 1999; Rozzi *et al.*, 1999) and mining (Ericsson and Hallmans, 1996).

The Palo Verde nuclear generating station near Phoenix, Arizona is the only commercial nuclear power station in the world that uses treated sewage effluent as a source of water for cooling tower operation (Blackson and Moreland, 1998). Treated effluent is pumped 58 km from the city of Phoenix WWTP to the onsite water reclamation facility, where it receives additional multistage biochemical treatment. During the summer months, 180 m^3 reclaimed water per minute is needed to compensate the evaporation losses in the cooling towers. In Harlingen, Texas, the development of an industrial park was subject to the availability of an affordable process water supply. As water resource options were very limited, the reuse of municipal wastewater was the only sustainable option and a 12500 m^3/day water reclamation facility was constructed. This has lead to the creation of over 3000 jobs in the textile plants in the industrial park (Filteau *et al.*, 1995). Although recycling and internal wastewater recuperation is common practice in many European industries, not many plants have resulted to reclaimed municipal wastewater for their process water. In the Netherlands, a large project is underway to assess the potential use of reclaimed WWTP effluent as an industrial process water in seven industrial parks in South-Limburg (Anonymous, 1999; Ruiters *et al.*, 1999). In Belgium, WWTP effluent is reused as a cooling and rinsing water in the waste incinerators of Bruges, Ostend and Wilrijk (Thoeye *et al.*, 2000).

4.4. Groundwater recharge

The use of groundwater recharge as a reuse application can satisfy multiple objectives: (1) arresting the decline of groundwater levels due to over-extraction, (2) protection of coastal aquifers against saltwater intrusion from the ocean and (3) storage of surplus water for future use (Asano and Levine, 1998). Groundwater aquifers provide natural mechanisms for treatment, storage and sub-surface transmission of reclaimed water. During soil aquifer treatment (SAT), or geopurification, a natural polishing effect of the treated wastewater is achieved, the key purification mechanisms being volatilisation, chemical or biological conversion, precipitation of metals and sorption to the soil (Asano and Levine, 1995; Oron, 1996). In addition, storing water underground has several advantages: (1) the cost may be lower than surface reservoirs, (2) the aquifer can serve as an eventual distribution system, (3) water stored in surface reservoirs is subject to evaporation, algae growth and pollution, (4) suitable reservoirs may not be available or environmentally acceptable, and (5) the inclusion of groundwater recharge in a reuse scheme may also provide psychological and aesthetic benefits as a result of the transition between reclaimed wastewater and groundwater. Reclaimed water is used to recharge groundwater either by surface spreading and subsequent percolation through soils, or by direct injection of treated wastewater into a groundwater aquifer. Of the greatest concern with using reclaimed water for groundwater recharge is contamination of the aquifer as the water extracted from the aquifer needs to be free of physical, chemical, microbiological radiological contaminants.

Good examples of this application can be found in Israel, where municipal effluent is used for agricultural irrigation after SAT (Kanarek *et al.*, 1993; Kanarek and Michail, 1996; Oron, 1996,1998). Many water reclamation facilities use SAT as an additional treatment step in the indirect production of potable water from reclaimed wastewater; more detail is given below.

4.5. Environmental and ecosystem restoration

In addition to providing a dependable, locally controlled water supply, water recycling offers tremendous environmental benefits. Four major advantages can be identified (US EPA, 1998): (1) decrease of diversion of freshwater from sensitive ecosystems, (2) decrease of wastewater discharge into sensitive water bodies, (3) creation or enhancement of wetlands and riparian habitats by use of reclaimed water and (4) reduction and prevention of pollution.

Some Californian water-recycling projects have been designed with explicit environmental and ecosystem benefits in mind (Gleick, 2000). For example, the development of water recycling projects in Los Angeles has provided a way to partially offset the loss of Mono basin water, while the implementation of the water reclamation facilities of San Francisco gave rise to a significant reduction of wastewater effluent discharge into the sensitive saltwater marsh ecosystem (US EPA, 1998). In Japan, tertiary treated effluent is used to augment and restore the flow of rivers in the Tokyo metropolitan area (Nakazato and Kawamura, 1997).

4.6. Non-potable urban reuse

Reclaimed wastewater can also be used to satisfy secondary water requirements in urban areas. Typically, urban reuse systems are derived from secondary treatment processes supplemented with sand filtration and high-dose chlorination (Asano and

Levine, 1998). The quality of such a reclaimed water is generally appropriate for unrestricted non-potable reuse, which could serve many uses, such as toilet flushing (commercial and residential buildings), fire protection, cleaning (vehicles, buildings, streets) and commercial air conditioning (Okun, 2000). In this respect, the development of dual distribution systems (one for high quality potable supply and one for reclaimed water) is a growing practice world-wide, particularly in areas with high rates of urban water usage. Because the cost of retrofitting reclaimed water pipes can render an entire project economically unfeasible, in most cases dual systems are installed as a part of a new development. It has been estimated that for new buildings over seven stories, the additional cost of providing a dual distribution system adds only 9 % to the cost of plumbing (US EPA, 1998). In dual distribution systems, the utmost care must be taken to prevent possible crossconnections between potable and non-potable lines. Some safety measures include: (1) complete segregation of both systems, (2) use of colour coded labels to distinguish potable and non-potable pipes, (3) backflow prevention devices, (4) periodic testing with tracer dyes to detect possible crossconnections and (5) irrigation during off hours to further minimise the potential for human contact (Asano and Levine, 1998).

Although the establishment of a dual distribution system for urban reuse can be traced back to the beginning of the 20th century (in 1926 a system was installed in Grand Canyon Village, Arizona), the municipality of St Petersburg, Florida is acknowledged to be the first city to implement a major comprehensive dual distributions system for residential, commercial and industrial customers. By 1995, nearly 8600 customers were connected, receiving 80000 m^3/day of reclaimed water. This program has halted potable water demand despite substantial population growth (Johnson and Parnell, 1998). In 1991, the Irvine ranch Water district was the first US water district to obtain permission for indoor reclaimed water use. Since then, the potable water demand has dropped 75 % in the seven facilities using reclaimed water for toilet and urinal flushing (Young *et al.*, 1998). England's first in-building water reclamation facility was situated in the Millennium Dome, Greenwich, where a mix of rainwater, groundwater and hand basin ("grey") water was reclaimed for toilet flushing (Khow *et al.*, 2000). Japan's water reuse is decisively directed towards non-potable urban applications. It is estimated that publicly owned WWTPs and on-site recycling systems annually provide $74*10^6$ m^3 of reclaimed water in more than 650 locations for toilet flushing (Ogoshi *et al.*, 2001). In Tokyo alone, approximately 2000 m^3/day of reclaimed wastewater is used in 23 high-rise buildings for this purpose (Nakazato and Kawamura, 1997).

4.7. Potable reuse

4.7.1. Indirect potable reuse

Indirect potable reuse encompasses the recovery of water from wastewater for the purposeful reintroduction into either a surface water or groundwater that ultimately serves as a drinking water supply. The primary incentives for indirect potable reuse are: (1) high cost and environmental impact of the conventional approach, (2) more stringent wastewater disposal standards, (3) water rights limitations (4) economic viability and (5) political viability (when water conservation measures are exhausted) (McEwen, 1998).

Some examples of indirect potable reuse facilities are summarised below:

- The Los Angeles County Sanitation District built the first large-scale water reclamation project of this type in the 1960s. One of the six reclamation plants in the area, the Whittier Narrows facility, discharges its tertiary treated effluent into spreading areas that recharge the groundwater, which serves as the potable water supply for some area residents. Health effect studies have shown that over the past three decades, the proper use of this reclaimed water, along with the employment of various safety guidelines, has not resulted in any health effects (Crook *et al.*, 1999; Hartling and Nellor, 1998).
- In Occoquan, northern Virginia, effluent from the 56000 m^3/day Manassas advanced treatment plant is discharged directly into the Occoquan reservoir, which is a principal drinking water resource for about 1 million people. No negative health effects attributable to the plant or the effluent discharges have been reported (McEwen, 1998).
- Since 1976, the Orange County California Water District's Water Factory 21 has been injecting up to 57000 m^3/day highly treated municipal wastewater into water supply aquifers. The recovery treatment train includes lime clarification, air stripping, recarbonation, filtration, carbon adsorption, reverse osmosis and disinfection. The injected water creates a freshwater mound or pressure ridge that hydraulically prevents saltwater intrusion, but a part flows inland and augments potable water supplies (Mills *et al.*, 1998).
- A major indirect potable reuse project is currently under construction in Wulpen, Belgium. Traditionally, the Intercommunal Water Company of Veurne-Ambacht extracted groundwater from the quaternary layer of the coastal dunes. However, the observation that further increase of groundwater extraction would inevitably result in saline intrusion has lead to the integration of wastewater

reclamation into the water cycle (Van Houtte *et al.*, 2000; Van Houtte *et al.*, 1998). An estimated $2.5*10^6$ m^3 of effluent will be treated annually by microfiltration and reverse osmosis and, after a residence time of 4 to 7 weeks in the unconfined aquifer in the dunes, reused for the production of potable water (Figure 2). Upon its commissioning in 2003, it will be the first full-scale indirect potable reuse facility in Europe. A similar project is being investigated at the Heist drinking water production facility (Thoeye *et al.*, 2000).

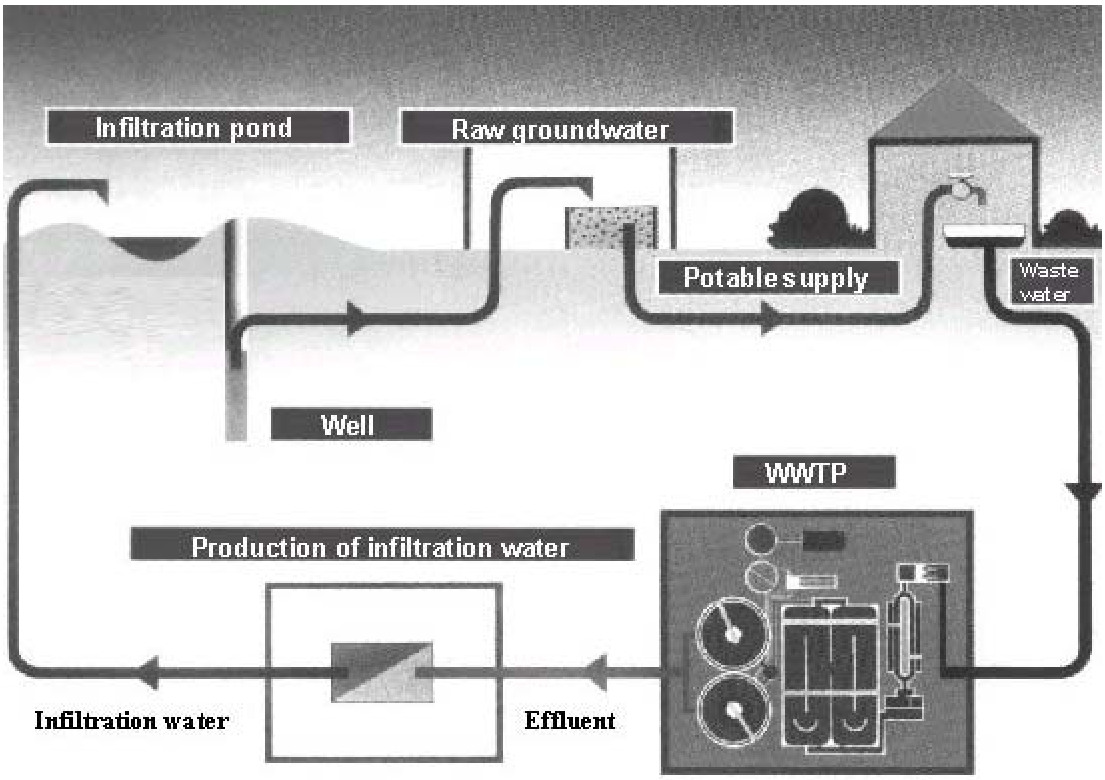

Figure 2 Integration of wastewater reclamation into the drinking water cycle in Wulpen, Belgium (Parmentier *et al.*, 2000)

4.7.2. Direct potable reuse

The most known, and to date, the only reuse system to directly produce potable water from wastewater is situated in Windhoek, Namibia. In 1969, the 4500 m³/day facility was commissioned and since 1971 the produced reclaimed water is blended with the available freshwater to a maximum of 30 %. After primary and secondary treatment, the water is impounded and when seasonally necessary, taken in and subjected to chemical clarification, activated carbon filtration, reverse osmosis, ozonation and chlorine disinfection (McEwen, 1998). Toxicological and epidemiological studies over more than 20 years showed no relationship between drinking water source and incidence of diarrheal diseases (Crook *et al.*, 1999). The plant is to be expanded to a capacity of 21 000 m³/day (Haarhoff and van der Merwe, 1996).

In the footsteps of this pioneering reuse plant, a number of extensive pilot and demonstration tests were undertaken to assess the health effects of direct potable reuse (*e.g.* Denver potable health effect pilot study, San Diego and Tampa studies). Although these studies convincingly demonstrate that an advantageous combination of treatment technologies can produce an effluent directly suited for drinking, none of these projects have resulted in full-scale applications (Crook *et al.*, 1999). In addition to public opposition, two major causes can be identified (Okun, 2000):

1. There have been no studies establishing possible long-term health effects. Moreover, at the moment it is beyond the capacity of epidemiology to establish the health effects of long-term exposure to trace amounts of contaminants, whether microbiological or chemical.
2. Potable water reuse is seldom necessary, as water conservation can for example be established via non-potable dual distribution systems. In other words, all other reuse resources should be exhausted prior to considering direct potable reuse.

Although it can be argued that the case of Windhoek contradicts these statements, the general consensus still is that "potable reuse, without the added protection provided by storage in the environment is a non-viable option" (Crook *et al.*, 1999).

5. RECLAIMED WATER QUALITY CONCERNS

A crucial presupposition to wastewater reuse is the reliable elimination of hazardous substances. Inconsiderate use of reclaimed water can have deleterious consequences - the most important being an adverse effect on the public health. However, also industrial processes and agricultural, urban and environmental applications may be negatively impacted. Therefore, a characterisation and discussion of the microbiological and chemical aspects of wastewater is warranted to provide an appreciation of the need to adopt and enforce criteria for the production, distribution and safe use of reclaimed water. Moreover, as some uses of reclaimed water demand a pristine appearance, organoleptic aspects are also taken into consideration.

5.1. Microbiological aspects

Where reclaimed water is used for applications that have potential human exposure routes, the major acute health risks are associated with exposure to biological agents, including bacterial pathogens, parasites (helminths and protozoa) and enteric viruses.

5.1.1. Occurrence of pathogens

The main source of pathogens in domestic wastewater is the faeces of infected individuals; thus municipal wastewater can contain a wide variety and concentration range of these agents. Typically, municipal wastewater contains hundreds of types of pathogenic micro-organisms, some of which are indicated in Table 1.

Table 1 Concentration range and infectious dose of pathogenic agents potentially present in raw domestic wastewater (adapted from Cooper and Olivieri (1998), Yates and Gerba (1998) and Crook (1998))

Agent	Concentration (number/100 mL)	Infectious dose	Disease
Bacteria	10^6 - 10^{12}		
Salmonella spp.	10^2 - 10^4	10^4 - 10^7 [a]	Salmonellosis
E. coli	10^5 - 10^7	10^6 - 10^{10}	Gastro-enteritis
Shigella	1 - 10^3	10 - 180	Shigellosis
Vibrio cholerae	10 - 10^4	10^3 - 10^7	Cholera
Clostridium perfringens	10^3 - 10^5	1 - 10^{10}	Perfringens
Enteric viruses	10^2 - 10^4	1 - 10	
Protozoa	10^3 -10^5		
Entamoeba histolytica	1 - 10	20	Amebiasis
Cryptosporidium parvum	10 - 10^3	1 - 10	Cryptosporidiosis
Giardia lamblia	10 - 10^4	< 10	Giardiasis
Helminths	10 - 10^3		
Ascarsis lumbricoides	1 - 10	1 - 10	Ascariasis

[a] Infectious dose for *Salmonella tiphy*, which causes typhoid fever

One of the most common bacterial pathogens is the genus *Salmonella*, which can cause disease in humans and animals. The most severe form of salmonellosis is typhoid fever, caused by *Salmonella tiphy*. *Vibrio cholerae* is the disease agent for cholera, which is not common in the developed countries, but still prevalent in other parts of the world. Other species that have been isolated from raw wastewater include

Shigella, *Clostridium*, *Mycobacterium*, *Escherichia coli* and *Pseudomonas.* The protozoan parasite *Entamoeba histolytica* is responsible for amoebic dysentery and amoebic hepatitis, but it has not been an important disease agent since the 1950s. On the contrary, giardiasis and cryptosporidiosis are emerging as major waterborne diseases. In fact, cryptosporidiosis can be fatal to immuno-compromised individuals. *Giardia* cysts and *Cryptosporidium* oocysts are much more difficult to inactivate by chlorination than bacteria and viruses and can survive in the open environment for extended periods of time (Venczel *et al.*, 1997). Helminthic parasites found in wastewater comprise the stomach worm *Ascaris lumbricoides*, the whipworm *Trichuris trichiura* and the tapeworms *Taenia saginata* and *Taenia solium*. The most important human enteric viruses are the enteroviruses (polio, echo and coxsackie), Norwalk virus, rotaviruses, reoviruses, caliciviruses, adenoviruses and hepatitis A virus (Crook, 1998).

5.1.2. Pathogen inactivation during wastewater treatment

Primary treatment does little to remove bacteria, viruses and protozoan parasites, although the large helminthic eggs can be reduced to some extent through straightforward sedimentation. Secondary activated sludge is relatively efficient in removing infectious agents; both sedimentation and aeration play a role in pathogen reduction. Oxidation ponds can, given sufficient retention times, achieve a considerable removal of enteric pathogens, especially helminthic eggs (Yates and Gerba, 1998). Some pathogen removal efficiencies of primary treatment and secondary activated sludge are indicated in Table 2.

Table 2 Typical percentual removal of micro-organisms (%) by conventional primary and secondary activated sludge treatment (Gleick, 2000).

Agent	Primary treatment	Secondary activated sludge
Salmonella	0 - 15	70 - 99
Shigella	15	80 - 90
Entamoeba histolytica	0 - 50	Limited
Giardia cysts	50 [a]	75 [a]
Helminth ova	50 - 98	Limited
Enteric viruses	Limited	75 - 99

[a] From Cooper and Olivieri (1998)

While these removals of pathogens might seem large, the initial concentrations are also large (Table 1). As a consequence, pathogens can persist in secondary effluent in high enough concentrations to pose a potential threat to human health. Hence, for many wastewater reuse applications, advanced (tertiary) treatment or disinfection may be required to further reduce the number of infectious agents.

5.1.3. Occurrence of infectious diseases (Crook, 1998)

Many epidemiological studies have been dedicated to microbiological aspects of wastewater reuse and reclamation. These have established the possibility of transmittance of infectious agents through pathways such as wastewater-contaminated drinking water supplies, crop irrigation using minimally-treated wastewater, contact of farm workers with poorly treated reclaimed irrigation water or inhalation of aerosols originating from spray irrigation sites using undisinfected wastewater. Indeed, during the 20th century, a number of major disease outbreaks have been linked to the use of poorly treated wastewater for irrigation purposes. In developing countries, where untreated or poorly treated wastewater is used for the irrigation of market crops, outbreaks of enteric diseases are still reported.

On the other hand, during the last decades there have been no confirmed cases of infectious disease resulting from reclaimed water use in the US. This evolution is due to the adopted practice of providing water of a quality appropriate for the highest use of reclaimed water in a community. This implies that in most cases all reclaimed water receives some form of tertiary treatment or disinfection to make it virtually pathogen free. Thus, it does not represent an unreasonable risk of infectious disease from infrequent, inadvertent ingestion. This is confirmed by various health effect assessments which have clearly demonstrated that such a level of treatment produces clean and safe effluents and that, given a well-considered multi-barrier treatment approach, the indirect and even the direct reuse of reclaimed municipal wastewater can be completely safe (Crook *et al.*, 1999).

5.2. Chemical aspects

Apart from possible inhalation of volatile organic components (VOCs), the health effects associated with the chemical components potentially present in municipal wastewater are generally not of major concern when the reclaimed water is not to be consumed or used for irrigation of food crops. However, health issues may arise when reclaimed water percolates into potable aquifers as a result of irrigation or other uses or when direct or indirect potable reuse is considered. Also, certain chemicals may generate concerns regarding the suitability of reclaimed water for industrial, environmental and irrigational purposes.

5.2.1. Inorganic chemicals

The concentration of inorganic constituents depends mainly on the nature of the water supply, the wastewater source and the degree of treatment provided. The presence of total dissolved solids (TDS), heavy metals, residual chlorine or nutrients can limit the applicability of reclaimed wastewater (Crook, 1998).

- Total dissolved solids: Typically, the TDS level increases by 300 - 400 mg/L for each cycle of domestic use and in many cases by higher values for each cycle of industrial use (Odendaal *et al.*, 1998). Consequently, reclaimed water can be inept for industrial or agricultural reuse purposes because excessive amounts of TDS can eventuate in scaling and corrosion or cause loss in yield and plant damage due to salt accumulation in the crop root zone.
- Heavy metals: Cadmium, molybdenum, copper, zinc and nickel can bioaccumulate in the food chain or exhibit toxicity towards fauna and flora at elevated levels. Hence, their presence in reclaimed water can preclude environmental or agricultural applications although generally, residual levels are acceptable for reclaimed water that has received at least secondary treatment.
- Residual chlorine is toxic to many aquatic organisms and must be removed prior to discharge in sensitive aquatic systems. Second, excessive residual chlorine causes severe plant damage if reclaimed water is sprayed directly on foliage. For urban uses, a chlorine residual is often desirable to prevent bacterial regrowth in pipes and mains and to minimise the health implications of inadvertent ingestion of reclaimed water (Asano and Levine, 1998).
- Nutrients: The presence of nutrients such as nitrogen, phosphorus and potassium is desirable for agricultural and landscape irrigation, since it lessens the need to apply synthetic fertilisers. However, application of excessive amounts can lead to the leaching of nitrogen into the soil, and may give rise to groundwater concentrations exceeding the drinking water limits. Discharging high amounts of nutrients into aquatic environments creates the potential for algae blooms, leading to odours, an unsightingly appearance and eutrophic conditions. Many industrial processes demand limited amounts of nutrients in reclaimed water to prevent algael growth or biofouling of process equipment (Asano and Levine, 1998).

5.2.2. Organic chemicals

Many of the major organic constituents in municipal wastewater, such as protein, and carbohydrate are readily biodegradable. As such, they fuel microbial growth and consume oxygen, which can render the water inappropriate for industrial or other uses. In addition, they hinder disinfection processes and can lead to acute or chronic health effects if the water is reclaimed for potable purposes. The biochemical oxygen demand (BOD) is used as a gross measure for biodegradable organics. On the other hand, a wide variety of organic components potentially present in municipal wastewater tend to resist biodegradation.

Total organic carbon (TOC), a surrogate parameter representing the organic constituents in a water sample, is often used to designate these components which can be of natural origin (*e.g.* humic substances, tannin and lignin) or comprise man-made synthetic organic chemicals (SOCs). It is estimated that around 65000 chemicals are used routinely in manufacturing, agricultural production and daily living (Chang *et al.*, 1998). Some of them have a rather poor biodegradability and it can be expected that at least a fraction may find its way into municipal wastewater. A number of these SOCs have been found to possess acute or chronic toxicity or cause adverse human health effects, even at extremely low concentrations. This has raised health concerns, especially where reclaimed water is used for potable applications or for the irrigation of food crops. With respect to the latter, however, it has been postulated that most SOCs are too large to pass through the semi-permeable membrane of plant roots (Crook, 1998). Indeed, there has been no unequivocal epidemiological evidence to demonstrate any harm caused by even one potentially toxic chemical present in irrigation-bound wastewater (Chang *et al.*, 1998).

Three categories of drinking water relevant SOCs, which have received considerable attention, are briefly discussed:

- Pharmaceutical products: recently, major concern emerged with respect to the fate of prescription and other commonly used pharmaceutical drugs, such as lipid regulation agents, antibiotics, antiseptics, analgesics, anti-epileptics and beta-blockers. A large number of pharmaceuticals have been detected in municipal wastewater, surface water and even drinking water (Bruchet *et al.*, 2001; Drewes *et al.*, 2001; Heberer *et al.*, 2001; Pinkston and Sedlak, 2001; Sacher *et al.*, 2001). The prime sources are veterinary and human use as many of the numerous active substances used to treat illnesses are excreted unmetabolised or as active metabolites. Moreover, improper disposal of expired medication can contribute to the observed contamination (Andreozzi *et al.*, 2001). Many of these constituents are unaffected by primary and secondary wastewater treatment and can be retrieved in the WWTP effluent in the µg/L-range (Table 3), which emphasises their importance as emerging pollutants for potable reuse applications. Because pharmaceuticals are designed to be biologically active, their residuals have a significant potential for a wide range of largely unexplored ecotoxicological consequences and potentially human health effects. Although the effects of these components at the concentrations at which they are observed in the environment are still under debate, a conservative approach would suggest their removal at least from aqueous streams destined for human use (Khan and Ongerth, 2001).

- Endocrine disruptors: the presence and effects of pseudo-estrogenic substances are becoming increasingly clear (Bruchet *et al.*, 2001; Tanghe, 1999). A large number of environmental contaminants, such as polychlorinated biphenyls, alkylphenols (originating from surfactants and detergents), bisphenol A and phthalates (released from plastics) seem to possess hormone-mimicking capacities, even at very low concentrations. In addition, the presence of steroid hormones from human and animal excretions (*e.g.* 17β-estradiol) and from contraceptives (*e.g.* 17β-ethynestradiol) in the aqueous environment can also adversely affect the endocrine system. Conventional activated sludge treatment results in a removal of more than 97 % of the most relevant xenobiotic endocrine disruptors, while approximately 25 to 45 % of the 17β-estradiol is removed (Goto and Kuribayashi, 2001; Bruchet *et al.*, 2001). Nevertheless, endocrine disrupting chemicals have been detected in WWTP effluents either analytically (Bruchet *et al.*, 2001; Goto and Kuribayashi, 2001; Witgens *et al.*, 2001) or their estrogenic properties have been demonstrated via bioassays (Gagné and Blaise, 1998; Routledge and Sumpter, 1996; Tanghe *et al.*, 1999). These pollutants are sufficiently potent to cause fish to be born half male and half female and are associated, at least to some extent, with the declining trends in male human reproductive fertility (Janssens *et al.*, 1997).
- NDMA or N-nitrosodimethylamine has been particularly disconcerting in locations where chlorinated WWTP effluent is used for indirect potable reuse. NDMA is classified as a probable human carcinogen and the US EPA has established a lifetime 10^{-6} cancer risk level of 0.7 ng/L (Najm and Trussell, 2001). A recent Californian regulations set an action level of 20 ng/L, which

resulted in the suspension of 2 drinking water production wells which were partly fed by recharge water from the wastewater reclamation system of the Orange County Water District's Water Factory 21 (Mitch and Sedlak, 2002). The occurrence in chlorinated, highly purified wastewaters while absent in the corresponding influent stream, suggests its formation as a possible DPB (Choi and Valentine, 2002)

Table 3 Pharmaceuticals observed in secondary WWTP effluents (Bruchet *et al.*, 2001; Drewes *et al.*, 2002; Heberer *et al.*, 2001; Pinkston and Sedlak, 2001)

Use	**Component**	**Concentration (ng/L)**
Antibiotics	Cyclamidomycin	Trace amount [a]
Analgesics/ anti-inflammatory	Diclofenac	50 [c] - 2500 [b]
	Ibuprofen	200 [c] - 3380 [d]
	Ketoprofen	45 [d] - 50 [c]
	Naxopren	100 [c] - 6280 [d]
Lipid regulating agents	Clofibric acid	480 [b]
	Gemfibrozil	1235 [d] - 1400 [c]
Anti-epileptics	Carbamazepine	1630 [b]
	Primidone	110 [d]
Beta-blockers	Metoprolol	250 [c]

[a] Bruchet *et al.* (2001) [b] Heberer *et al.* (2001)
[c] Pinkston and Sedlak (2001) [d] Drewes *et al.* (2002)

5.3. Organoleptic aspects

Aesthetic aspects of water designated for reuse applications are mainly associated with the presence of turbidity, colour, odour and particulates, although the presence of latter can have more than aesthetical implications.

- Suspended solids: Particulates can adsorb organic constituents, heavy metals and shield micro-organisms from the impact of disinfectants, especially if UV disinfection is applied. In addition, suspended matter can reduce the efficiency of chlorine or ozone disinfection through reaction with the disinfectants or lead to sludge deposits and anaerobic conditions when disposed into aquatic environments. Excessive suspended solids concentrations can cause clogging of process water pipelines, sprinklers and especially drip irrigation systems.
- Colour, odour, turbidity: High level uses, *e.g.* urban irrigation or toilet flushing, require water that should be no different in appearance than potable water, *i.e.* colourless, odourless and clear.

6. WATER RECLAMATION AND REUSE CRITERIA

The protection of public health is a prerequisite for any wastewater reclamation and reuse application. For most non-potable reclamation purposes, health effects are predominantly associated with microbiological agents, but health effects related to organic constituents must also be considered when reclaimed water is used for potable purposes. Because the extent of direct exposure is a decisive health risk factor, many regulatory agencies stipulate water quality standards that are coupled to the specific application of the reclaimed wastewater. Second, since health risks are also associated with the adequacy, effectiveness and reliability of the treatment system, these criteria are often linked to requirements for treatment, sampling and monitoring. In addition, quality criteria often contain guidelines to ensure optimal process performance (by minimising phenomena such as scaling, corrosion, fouling clogging,...) and to minimise secondary adverse effects of reuse.

6.1. United States

In 1992, the United States Environmental Protection Agency, in conjunction with the U.S. Agency for International Development, issued “Guidelines for Water Reuse” (US EPA, 1992). Because specific reuse criteria already had been developed in individual states, the primary purpose was to provide guidelines with supporting information for utilities and regulatory agencies, particularly in states where standards did not exist or were being revised or expanded. The guidelines address all important aspects of water reuse and include recommended treatment processes, reclaimed water quality limits, monitoring frequencies, setback distances and other controls for various water reuse applications. A summarised version of the regulations is reported in Table 4.

The fact that the guidelines stipulate both quality criteria and treatment is based on the following observations (Crook, 1998): (1) characterisation of reclaimed water by surrogate parameters alone is insufficient, (2) a combination of treatment type and quality requirements known to produce a particular reclaimed water quality can preclude the need to monitor the finished water for certain contaminants, (3) expensive, time-consuming and in certain cases questionable monitoring of pathogenic micro-organisms can be eliminated without compromising the public health protection and (4) the treatment reliability is enhanced. Notably, it is advised that, regardless of the water use, some level of disinfection should be provided to avoid adverse health effects from inadvertent contact, accidental or intentional misuse of a water reuse system.

Table 4 EPA suggested guidelines for reuse of municipal wastewater (US EPA, 1992)

Type of reuse	Treatment	Reclaimed water quality	Comments
• Urban reuse • Agricultural reuse [a] • Recreational impoundments	• Secondary • Filtration • Disinfection	pH = 6-9 $BOD_5 \leq 10$ mg O_2/L NTU ≤ 2 [b] FC ≤ 1 / 100 mL [c] res. $Cl_2 \geq 1$ mg/L [d]	Reclaimed water should be clear, odourless and contain no substances, toxic upon ingestion
• Restricted irrigation • Agricultural reuse • Industrial reuse • Environmental reuse	• Secondary • Disinfection	pH = 6-9 $BOD_5 \leq 30$ mg O_2/L SS ≤ 30 mg/L FC ≤ 200 /100 mL [e] res. $Cl_2 \geq 1$ mg/L [d]	Minimise contact with reclaimed water
• Groundwater recharge	• Site and use specific • Primary (min. spreading) • Secondary (min. injection)	Site and use specific	Facility should be designed to ensure that no reclaimed water reaches potable supply aquifers
• Indirect potable reuse *By spreading*	• Site-specific • Secondary and disinfection (min.)	Site specific Meet drinking water standards after percolation	Reclaimed water should be under-ground for at least 1 year
• Indirect potable reuse *By injection* *By surface water augmentation*	• Primary • Secondary • Disinfection • Tertiary treatment	pH = 6-9 FC ≤ 1 / 100 mL [c] NTU ≤ 2 [b] res. $Cl_2 \geq 1$ mg/L [d] Drinking water standards	Reclaimed water should be under-ground for at least 1 year Additional quality standards apply

[a] For food crops that are not commercially processed, *i.e.* those that, prior to sale to the public or others, do not receive chemical or physical processing sufficient to destroy pathogens

[b] Prior to disinfection. Average over 24 hours; should not exceed 5 NTU. If SS is used in stead of turbidity, the average SS should not exceed 5 mg/L

[c] Faecal coliform median value over 7 days. Should never exceed 14 / 100 mL

[d] Total chlorine residual after a minimal contact time of 30 minutes. Dechlorination may be necessary prior to environmental applications

[e] Median value over 7 days. Should never exceed 800 / 100 mL

6.2. World Health Organisation

The WHO guidelines, introduced in 1989, are primarily intended as water quality criteria for agricultural applications in developing countries. As such, they are based on the observation that the main health effects are to be associated with helminthic diseases. It is reasoned that, if the guidelines of Table 5 are met, other pathogens such as trematode eggs and protozoan cyst are also reduced to undetectable levels. Although supported by epidemiological evidence, the guidelines as a whole are meant to provide a design standard or to introduce at least some form of treatment prior to crop irrigation.

Table 5 World Health Organisation microbiological health guidelines for wastewater reuse in agriculture (WHO, 1989) [e]

	Reuse conditions	**Exposed groups**	**Intestinal nematodes**[a]	**Faecal Coliforms**	**Technological requirements**
A	Unrestricted irrigation *Crops likely to be eaten uncooked, sports fields, public parks* [c]	Workers Consumers Public	≤ 1 egg/L [b]	≤ 10^3 / 100 mL [b]	Series of stabilisation ponds or equivalent treatment
B	Restricted irrigation *Cereal crops, industrial and fodder crops, pasture and trees*	Workers	≤ 1 egg/L [b]	No standard recommended	8 -10 days of retention in stabilisation pond or equivalent
C	Localised irrigation *Crops as in B if exposure to workers and the public does not occur*	None	Not applicable	Not applicable	As required by irrigation technology, no less than primary sedimentation

[a] *Ascaris* and *Trichuris* species and hookworms

[b] During the irrigation period

[c] A more stringent guideline (200/100 mL)applies to public lawn, with which the public may come into contact

[d] For fruit trees, irrigation should cease 2 weeks before fruit is picked

[e] In specific case, local epidemiological, socio-cultural and an environmental factors should be taken into account and the guidelines modified accordingly

They can be considered appropriate as an interim measure in developing countries until higher quality effluent can be produced. Recently, Blumenthal *et al.* (2000) published recommendations for revising the WHO guidelines, based on epidemiological, microbiological and risk assessment studies published since the 1989 WHO guidelines. A lower nematode guideline of 0.1 egg/L is proposed, based on epidemiological data involving surface irrigation, lower temperatures or exposure to children. Also, standards for faecal coliforms of 10^5/100mL (spray irrigation) and 10^3/100mL (flood irrigation or contact with children) are suggested for restricted irrigation.

6.3. Japan

The water quality criteria for toilet flushing, the hallmark of Japanese wastewater reclamation and reuse, have been established in 1981 by the Ministries of Health and Welfare, International Trade and Industry, and Construction. The latter also developed the guidelines for landscape irrigation and environmental water. Table 6 displays the various water reclamation standards.

Table 6 Japanese water reclamation criteria (Ogoshi *et al.*, 2001)

Parameter	Toilet flushing	Landscape irrigation	Environmental water
Total coliforms (CFU/mL)	≤ 10	Not detected	Not detected
Residual chlorine (mg/L)	Trace amount	≥ 0.4	-
Turbidity (NTU)	-	-	≤ 10
BOD_5 (mg O_2/L)	-	-	≤ 10
pH	5.8 - 8.6	5.8 - 8.6	5.8 - 8.6
Appearance	Not unpleasant	Not unpleasant	Not unpleasant
Odour	Not unpleasant	Not unpleasant	Not unpleasant

6.4. European Union

The legal status of water reclamation of reuse varies considerably throughout the European Union (Bontoux, 1998). France has some national recommendations, Italy has a national law complemented by technical standards for wastewater reuse in agriculture and in Spain, various autonomous regions have developed their own quality guidelines. In Greece and Portugal, national standards are under consideration, while in many other European countries, the regulatory status of wastewater reclamation is non-existent.

On a European level, the only regulation so far is article 12 of the European Wastewater Directive 91/271/EEC (European Commission, 1991), which stipulates that *"treated wastewater shall be reused whenever appropriate"*. Apparently, this has been adopted in the most recent version of the Flemish environmental legislation (VLAREM II, 2001), as the statement in Article 5.3.1.4. §1 *"Gezuiverd afvalwater dient indien mogelijk te worden hergebruikt"* is the translation of the above. The recent water framework directive (European Commission, 2000), which has to be converted into the individual member states' legislation, stipulates reuse as one of the possible tools to prevent deterioration of the water bodies. Thus, the exact incorporation of reuse and reclamation into future water management and water supply guidelines is left to the member states.

6.5. Prospects for international guidelines for water reclamation

Recently, a concept of international guidelines for wastewater reclamation was developed by an international panel of scientists (Anderson, 2001; Anderson *et al.*, 2001). As it is recognised that microbiological risks are usually the dominant risk for non-potable reuse, there are basically 2 approaches to health risk assessment:

- *Quantitative microbiological risk assessment*, which is basically the theoretical calculation of a small risk to which the community is exposed by particular practices, *e.g.* water reuse. It allows the determination of extremely low risks, which will never be statistically demonstrable in a community where recycling occurs. This high technology / high cost / low risk approach is exemplified in the Californian regulations.
- *Attributable risk.* This is a risk derived from epidemiological studies, and guidelines derived thereof are based on the principle of incurring no incremental risk on the population. This low technology / low cost / high risk approach is embodied in the 1989 WHO regulations.

Despite these different approaches, the risks derived from a particular use of a particular reclaimed water are universal. With this in mind, an international framework with a series of steps progressing from low quality / high risk to high quality / low risk as shown in Table 7 was proposed. Other requirements, such as additional microbiological or chemical guidelines, may be combined with this star grading. Because "tolerable" risks are conditioned by local circumstances, public acceptability and affordability, choosing a step on the quality/risk ladder is to be based on the national / provincial circumstances. For example, a developed country may deem a

4-star rated water for irrigation of non-food crops necessary, while a developing country may judge a 3-star rated water of sufficient quality (Think globally, act locally).

Table 7 Possible recycled water grades and treatment. Key: UASB = upflow anaerobic sludge blanket reactor (Anderson *et al.*, 2001)

Grade		Description	FC/100 mL	Treatment
	5 star	Potable		Advanced multi-barrier treatment
A^+	4 star	Open Access	< 1	Secondary + membranes (or coagulation / filtration) + disinfection
A	3 ½ star	High contact	< 10	Secondary + filtration + disinfection
B	3 star	Medium contact	< 100	Secondary + disinfection
C	2 star	Low contact	< 1000	Secondary (or UASB) + disinfection
D	1 star	Restricted	< 10000	Secondary + maturation pond

Hence, the creation of an international framework combining the high technology / high cost / low risk with the low technology / low cost / high risk approach, could provide benefits towards better risk management, a better balance between risk and affordability, greater public understanding, better focussed research and development and greater project certainty.

CHAPTER 3

WASTEWATER RECLAMATION TECHNOLOGY

1. GENERAL OVERVIEW

The effective treatment of wastewater to meet water quality objectives for water reuse applications and to protect public health is a critical element of water reuse systems. A generalised view of the major technologies that are available for wastewater treatment and wastewater reuse and recycling systems is given in Figure 1.

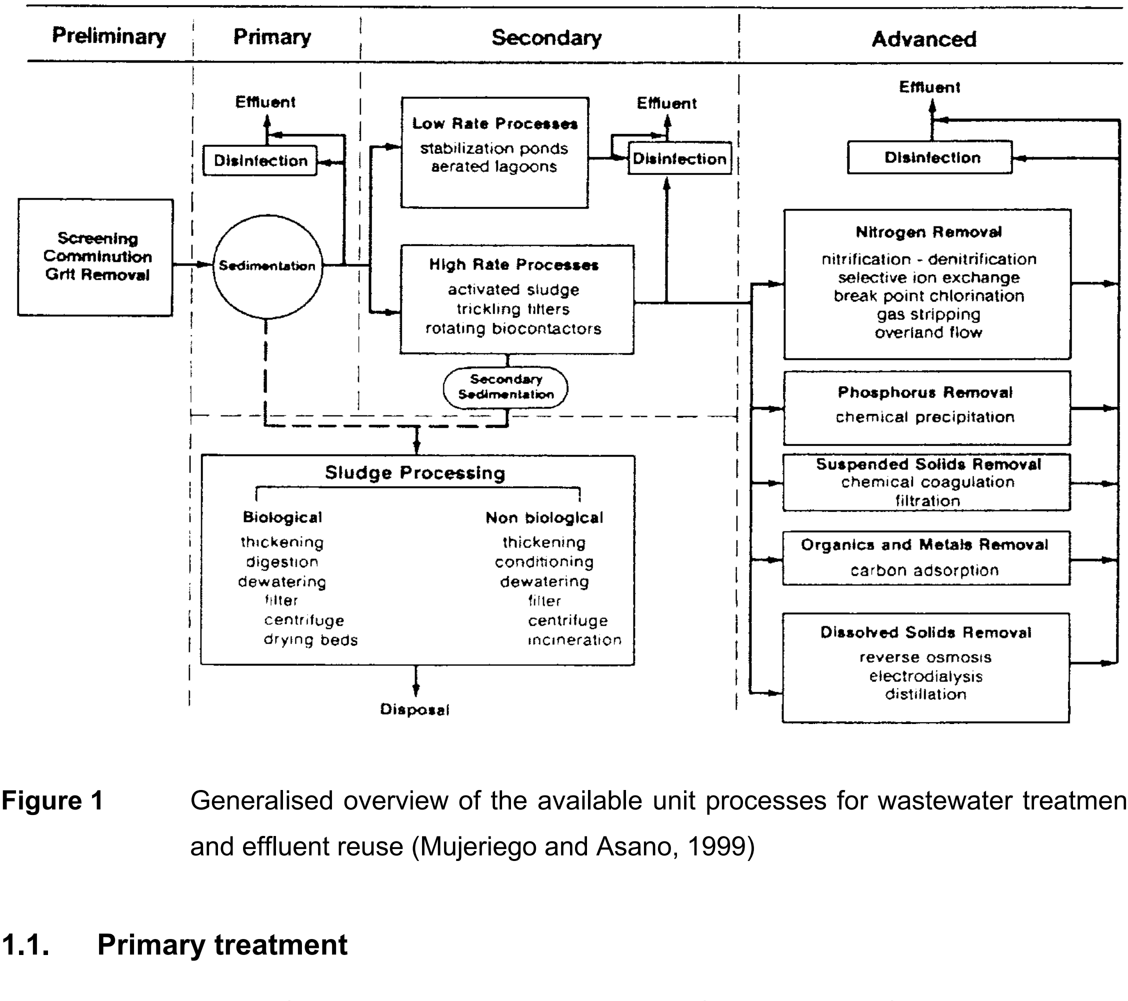

Figure 1 Generalised overview of the available unit processes for wastewater treatment and effluent reuse (Mujeriego and Asano, 1999)

1.1. Primary treatment

Primary treatment refers to the initial processing of wastewater for the removal of settable organic and inorganic solids as well as floating materials. In conventional wastewater treatment schemes, this is achieved through screening and comminution for the removal of large solids, grit removal and sedimentation, while floating materials (such as oil and grease) are eliminated through skimming. In general, particles larger than 50 µm, around 50 % of the suspended solids and 25 to 50 % of the BOD can be removed by these processes (Metcalf & Eddy, 1991). For most reuse applications, primary treatment alone is insufficient to meet water quality objectives.

1.2. Secondary treatment

Secondary treatment systems include a variety of biological treatment processes coupled to solid/liquid separation. In the biological process, a part of the degradable dissolved and suspended organic matter in the wastewater provides energy for microbial growth, while the remainder is microbially oxidised to water, carbon dioxide and other end products. The microbial biomass can be suspended (*e.g.* activated sludge, stabilisation ponds and aerated lagoons), but can also be fixed in biofilms (*e.g.* trickling filters and rotating biological contactors). After solid/liquid separation, where the excess biosolids are removed, the effluent typically contains levels of BOD and suspended solids ranging from 10 - 30 mg/L. Depending on the process operation, 10 to 50 % of the organic nitrogen can be removed and phosphorus is converted to phosphate. For some reuse applications, the quality of secondary effluent can be sufficient; typical applications include agricultural irrigation (Hussain and Al-Saati, 1999) and groundwater recharge (Kanarek and Michail, 1996; Oron, 1996).

1.3. Tertiary or advanced treatment

Typically these processes are performed downstream of conventional biological treatment processes (*e.g.* activated sludge, trickling filters, waste stabilisation ponds) to remove additional dissolved and suspended contaminants, nutrients, specific metals, and other harmful constituents (Mujeriego and Asano, 1999). A more detailed discussion is provided below.

1.4. Disinfection

Disinfection is an essential treatment component for almost all wastewater reclamation and reuse applications. The objective of disinfection is to protect human health by destroying waterborne pathogenic micro-organisms. A number of disinfection technologies can be distinguished (Pilkington, 1995):

- Chlorination is the oldest, least expensive and most commonly used disinfection agent used in wastewater reclamation. Typical doses range from 5 to 20 mg/L with a maximum contact time of two hours. Because residual chlorine can have adverse effects on irrigated crops and aquatic fauna and flora, dechlorination (by means of sulphur dioxide or activated carbon) is sometimes applied prior to use.

- Other chemical disinfectants include chloramine, bromine, chlorine dioxide, ozone, hydrogen peroxide, iodine and silver ions. Some must be generated *in situ,* some may leave no residual, some may liberate undesired by-products and others are prohibitively expensive.
- Physical disinfection by ultraviolet (UV) irradiation has earned a reputation as a viable alternative to chemical disinfection processes in wastewater reclamation (Braunstein *et al.*, 1996). Alternatively, membrane filtration systems can be designed for micro-organism removal (Madaeni, 1999). Other physical processes, such as heat, ultrasound and irradiation are rarely used.

2. ROLE OF ADVANCED TREATMENT IN WASTEWATER RECLAMATION

Municipal effluent, treated to the point required by regulatory agencies, can satisfy some reuse applications, such as agricultural irrigation, but often tertiary or advanced treatment is needed to further augment its quality. As the quality of treated wastewater approaches that of unpolluted natural water, the concept of municipal wastewater and reuse is generated. In the extreme case, advanced treatment can even produce a water of much higher quality than drinking water; the product thus obtained is designated *repurified water* (Mujeriego and Asano, 1999). The concept of water quality changes taking place during drinking water production, municipal use and treatment and wastewater reclamation is visualised in Figure 2.

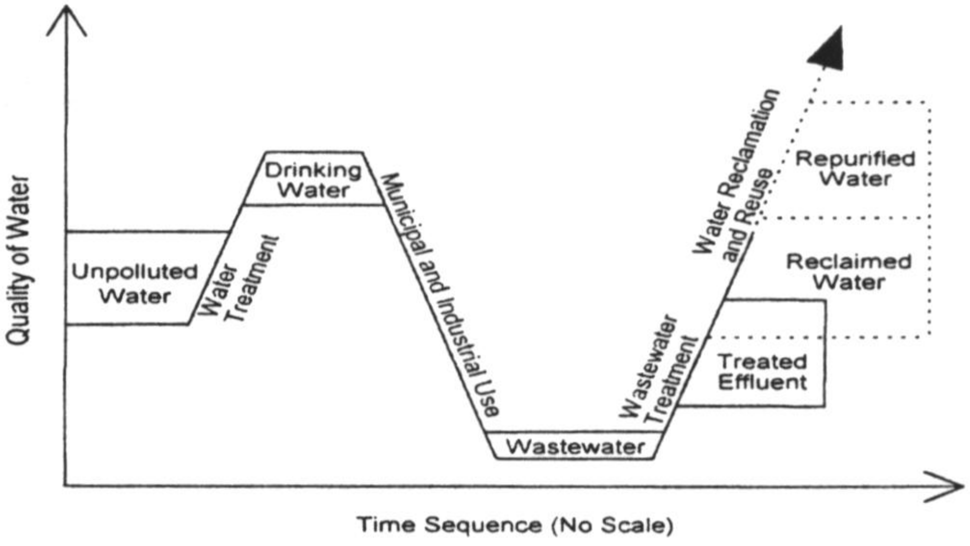

Figure 2 Water quality changes during municipal use of water, tertiary treatment and water reuse and reclamation (Mujeriego and Asano, 1999)

3. UNIT PROCESSES IN ADVANCED WASTEWATER TREATMENT

A large armada of advanced treatment technologies exists. Table 1 shows a classification of some unit processes used in wastewater reclamation based on their ability to treat five basic categories of contaminants (Zinkus *et al.*, 1998):

- *Inorganics* are generally those compounds not possessing carbon-hydrogen bonds. They include elemental ions, transition metal complexes and various hydrides of metalloids and non-metals.
- *Organics* include aliphatic and aromatic hydrocarbons, alcohols, aldehydes, ketones, ethers, carboxylic acids, esters, amides and complex organics such as pesticides, lignin, pharmaceuticals and so on.
- *Dissolved components* are smaller than colloidal particles (about 0.1 µm) and are dispersed as individual molecules or ions throughout the water.
- *Suspended solids* are particles larger than 0.1 µm, which can be filtered from the solution. Particles of sufficient size and density are often settlable; those that remain in suspension in a quiescent solution (*e.g.* colloids) are termed non-settlable.
- *Biological constituents* encompass bacteria, fungi, algae, protozoa and viruses.

Table 1 Applicability of advanced treatment technologies for different types of contaminants (Mujeriego and Asano, 1999; Zinkus *et al.*, 1998)

Technology	Inorganic	Organic	Dissolved	Suspended	Biological
Biological treatment	✓	✓	✓	✓	
Activated carbon		✓	✓		
Centrifugal separation				✓	
Chemical oxidation	✓	✓	✓		✓
Coagulation / flocculation	✓	✓		✓	
Evaporation	✓	✓	✓	✓	✓
Filtration	✓	✓		✓	
Flotation	✓	✓		✓	
Ion exchange	✓	✓	✓		
Membrane separation	✓[a]	✓	✓[a]	✓	✓[a]
Precipitation	✓	✓	✓	✓	
Stripping	✓	✓	✓		

[a] depending on membrane type and pore size

A short description of some of these unit processes is given below; membrane processes, evaporation and electrochemical treatment will be discussed separately:

- Biological treatment: after secondary biological treatment, advanced bio-technological processes can be implemented to further reduce the amounts of organics and mineral nitrogen. Recent examples include biological activated carbon oxidative filtration and novel oxidative cometabolic transformation pathways (Miserez *et al.*, 1999).
- Activated carbon is often used for removing hydrophobic organic compounds from municipal wastewaters. The treatment efficacy is contingent on many factors, including the amenability of the dissolved constituents to sorption, the presence of other substances which may enhance or impede the adsorption process, the soundness of engineering and proper operation and maintenance of the system (Ford, 1981). When coupled to solid/liquid filtration, AC can serve a dual role as tertiary filter and adsorber of organics (Kuo *et al.*, 1998). In biological activated carbon filters, these processes are further supplemented by biodegradation (Miserez *et al.*, 1999; Sirotkin *et al.*, 2001; van der Hoek *et al.*, 1999). A disadvantage of carbon adsorption is that it requires periodic regeneration (by means of solvent extraction or stream, thermal or chemical treatment) (Ford, 1981).
- Chemical oxidation: encompasses the partial or complete mineralisation of organic components into water and carbon dioxide, although also inorganic constituents (such as TAN, cyanides and sulphides) can be chemically oxidised. Common oxidants used include ozone, chlorine, chlorine dioxide, hydrogen peroxide, potassium permanganate and supercritical water. Advanced oxidation processes, such as Fenton oxidation, result in the (photo)catalytic production of hydroxyl radicals that oxidise the contaminants. Though generally more expensive than biological treatment, chemical oxidation processes have been cost-efficiently implemented in various reuse applications (Li and Zhao, 1999; Vandevivere *et al.*, 1998; Weber and LeBoeuf, 1999).
- Coagulation/flocculation : Coagulation comprises the addition of inorganic salts (aluminium sulphate, ferric chloride) which react with the particle surface resulting in particle destabilisation. Subsequently, flocculation is performed to aggregate particles into flocs that are susceptible to removal by gravitational clarification or filtration. In many municipal reuse treatment schemes, coagulation/flocculation is carried out directly after secondary treatment in order to remove excess residual solids (Madireddi *et al.*, 1997), or as a pre-treatment prior to membrane filtration (Abdessemed *et al.*, 2000; Speth *et al.*, 2000).

- Filtration: granular medium filtration is effective in removing suspended particles larger than 3 µm. It is stipulated as a required treatment process in many reuse applications to remove suspended solids that can compromise disinfection effectiveness, or as a pre-treatment to reduce the particulate loading on downstream reclamation processes (Asano and Levine, 1998).
- Ion exchange is the reversible exchange of ions between an insoluble solid salt (the ion-exchange resin) and a liquid in contact with that solid. It is very effective in separating low concentrations of dissolved inorganic species and organics from dilute wastewaters (Lin and Lin, 1998; Zinkus *et al.*, 1998).
- Precipitation is a process by which the chemical equilibrium of a solution is altered to reduce the solubility of the constituents of concern. Lime clarification, where the change in pH results in the precipitation of a number of inorganic constituents (such as calcium), as well a partial removal of biological and organic constituents, is sometimes used in advanced municipal wastewater treatment trains (McEwen, 1998).
- Stripping is a mass-transfer process in which air or steam is passed, generally countercurrently, through a waste stream. It is typically used for the removal of dissolved inorganics (TAN, hydrogen sulphide, hydrogen cyanide) and dissolved organics (VOCs), while non-volatiles will be unaffected (Lin and Lin, 1998; Zinkus *et al.*, 1998).

4. MEMBRANE FILTRATION PROCESSES

4.1. Overview

Among the advanced treatment processes, membrane applications have clearly emerged as one of the promising alternatives to conventional advanced physico-chemical treatment. Membrane processes employ a semi-permeable membrane and a driving force across the membrane to separate target constituents from a feed liquid solution:

- A *semi-permeable membrane* can be defined as an interphase that separates two phases and restricts the transport of various species in a rather specific manner (Madaeni, 1999).
- The *driving force* can be a pressure gradient , a concentration gradient (*e.g.* dialysis, liquid membranes), a temperature difference (*e.g.* membrane distillation, pervaporation) or an electrical potential (*e.g.* electrodialysis).

Especially the pressure-driven membrane processes are finding increased use in cost-comparable applications for the removal of micro-organisms, trace organic substances, specific ions and suspended and dissolved solids in advanced wastewater treatment and reclamation. Different types are used (Bilstad, 1997; Fane, 1996) :

- Microfiltration (MF) and ultrafiltration (UF) retain constituents according to their size and molecular mass and are generally defined by their nominal pore size or molecular cut-off weight (*i.e.* the molecular weight of the molecule that is retained for 90 %).
- Nanofiltration (NF) rejection is based both on sieving of smaller organic molecules and Donnan exclusion of multivalent ions.
- Reverse osmosis (RO) membranes are so dense that discrete pores do not exist and transport occurs via statistically distributed free volume areas (diffusion-solution mechanism).
- Hybrid processes: the coupling of one or more membrane processes to another unit process such as adsorption, coagulation or biological treatment, results in a hydrid process with a better performance than either of the component parts.

In Table 2, a more detailed classification of pressure driven membrane processes and their hybrid derivatives in terms of contaminant removal is given.

Table 2 Overview of membrane applications by contaminant type. Key: H1 = biological treatment + UF/MF; H2 = adsorption + UF/MF; H3 = chemical treatment + MF/UF; ± = partial removal / dependent on membrane type (Fane, 1996; Zinkus *et al.*, 1998)

Species	**Type**	**Size**	**MF**	**UF**	**NF**	**RO**	**H1**	**H2**	**H3**
Suspended	Particulates	> 1 µm	✓	✓	✓	✓	✓	✓	✓
	Colloids	0.1 - 1 µm	±	✓	✓	✓	±	±	±
Biological	Protozoa	> 10 µm	✓	✓	✓	✓	✓	✓	✓
	Bacteria	~ 1 µm	✓	✓	✓	✓	✓	✓	✓
	Cysts/oocysts	~ 0.1 µm	±	✓	✓	✓	±	±	±
	Viruses	0.01 - 0.1 µm		±	✓	✓	±	±	±
Organics	High MW	> 10 kD	±	✓	✓	✓			✓
	Middle MW	1 - 10 kD		±	✓	✓		✓	✓
	Low MW	< 1 kD			±	✓		✓	✓
Inorganics	Ions	< 0.1 kD			±	✓	✓		

As a rule of thumb, these processes remove turbidity and bacteria (MF), viruses and macromolecules (UF), small molecules and hardness (NF) and dissolved ionic species (RO) (Schafer *et al.*, 2000). Figure 3 compares the average pore diameter of conventional filtration, MF, UF and RO to the relative size of some solutes.

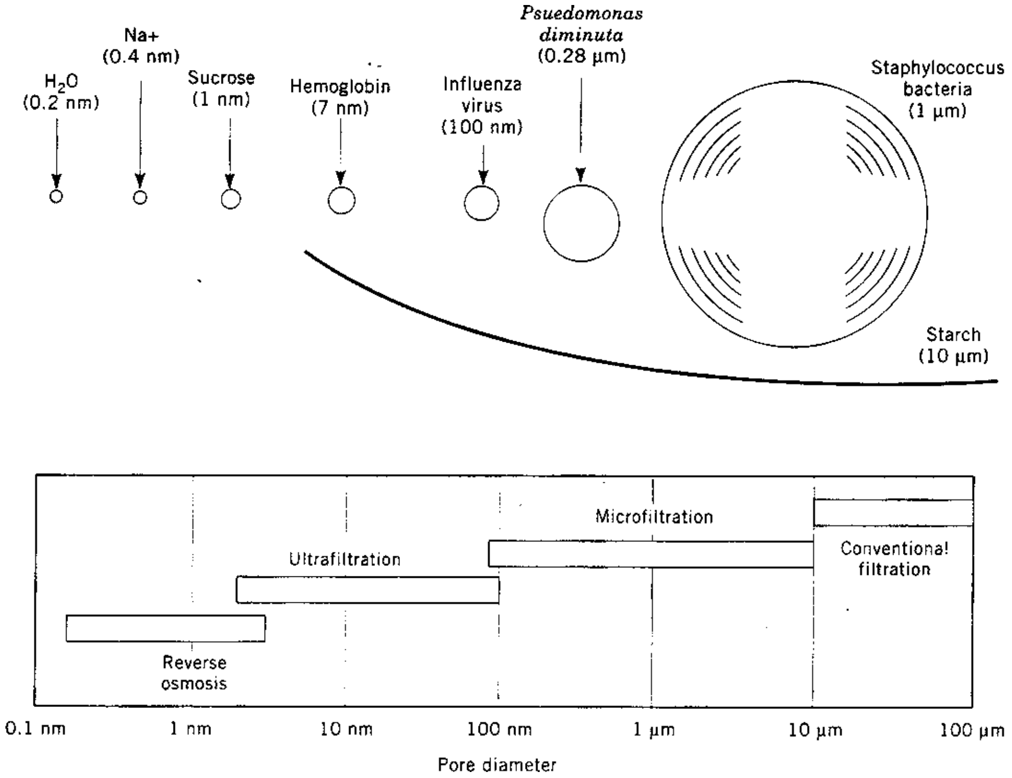

Figure 3 Effective ranges of RO, UF, MF and conventional filtration next to the relative size of different solutes removed by each class of membrane (Baker, 1998)

4.2. Membrane technology in municipal wastewater reclamation

4.2.1. Microfiltration, ultrafiltration

MF and UF downstream of secondary wastewater treatment has been described extensively in literature. The overall treatment produces a permeate suited for a wide variety of reuse applications, such as toilet flushing (Ahn and Song, 1999,2000), industrial process water (Tchobanoglous *et al.*, 1998; van Hoof *et al.*, 1998) and landscape irrigation (Jolis *et al.*, 1999). Because of the high permeate quality, MF and UF are often used as a pre-treatment prior to further desalination using reverse osmosis (van der Hoek *et al.*, 2000; van Hoof *et al.*, 1999; Van Houtte *et al.*, 2000).

These studies have demonstrated that direct processing of WWTP effluent can be both technically and economically feasible: the operational costs, including interest (7 %)

and depreciation (15 years), maintenance (3 % of investment annually), chemicals and electricity (0.05 EUR/kWh) are in the range of 0.09 - 0.22 EUR/m^3, depending on scale, operating procedure and feed water quality (van Hoof *et al.*, 1998).

4.2.2. Nanofiltration

Direct NF of WWTP effluent has been considered: Duin *et al.* (2000) found that nanofiltered effluent was not only superior in quality, but also much less susceptible to variations in influent composition than UF permeate. However, these advantages were offset by the considerably higher operational costs (0.30 - 0.50 EUR/m^3), leading to the conclusion that the choice of direct NF would have to be based mainly on quality considerations.

4.2.3. Reverse osmosis

The application of RO technology as a technology for municipal wastewater reclamation dates back to the early stages of commercialisation of the RO process. Because of the very high fouling potential of the feed stream, pre-treatment to remove colloidal and solid particles and to arrest biological activity, is essential (Wilf, 1998). This is conventionally achieved by a combination of coagulation/flocculation, lime treatment and granular medium filtration (Abdel-Jawad *et al.*, 1999; van der Hoek *et al.*, 2000), although MF/UF based pre-treatment has been demonstrated to produce a superior RO feed water quality (van Hoof *et al.*, 1999). RO permeate is employed in high-quality reuse applications, such as groundwater recharge, industrial process water and indirect potable reuse. Examples of pilot-scale and full-scale installations are provided in Chapter 2 (§ 4.7), by Reith and Birkenhead (1998) and by McEwen (1998).

Including pre-treatment, the total operational cost of full-scale installations can be as low as 0.34 EUR/m^3 (Abdel-Jawad *et al.*, 1999), but typically ranges from 0.5 - 0.7 EUR/m^3 RO permeate. For the planned WWTP effluent reclamation facility of Wulpen, Belgium, the projected permeate cost is 0.66 EUR/m^3 (Van Houtte, 1999). Recently Downing *et al.* (2002) showed that a ponding system supplemented by dissolved air flotation, slow sand filtration and disinfection could produce a comparable quality of RO feed water as activated sludge and MF at a considerably lower cost. The overall operational cost amounted to 0.77 EUR/m^3 for the ponding system, while for the activated sludge treatment train a cost of 1.40 EUR/m^3 was calculated. The difference was almost entirely due to the lower capital and energy expenditures of the ponding system.

4.2.4. Hybrid processes

Membranes can be coupled to biological treatment processes for biosolids separation (Husain and Côté, 1999; van der Roest *et al.*, 2001), bubbleless aeration of the biomass (Casey *et al.*, 1999) and extraction of selective pollutants (Livingston *et al.*, 1998). Biosolids separation is, however, the most widely studied and has found full-scale applications in many countries. Extensive reviews on this process, termed membrane bioreactor (MBR), have been presented by Gander *et al.* (2000a) and Visvanathan *et al.* (2000). The advantageous configuration of MBRs gives rise to a lower sludge production and a high treated water quality (Ghyoot and Verstraete, 2000). As a result, the permeate is suited for direct reuse in applications such as groundwater recharge or unrestricted non-potable reuse (Cicek *et al.*, 1998; Gander *et al.*, 2000b; Jefferson *et al.*, 2000). Compared to conventional activated sludge and MF, supplementing a MBR with RO has the additional advantages of low sludge production and absence of secondary waste streams. On the other hand, it is not always possible (or economical) to retrofit a WWTP with MBR technology (Côté *et al.*, 1997).

The hybrid process resulting from the addition of activated carbon to membrane filtration reactors is a well-recognised technology and full-scale applications in drinking water production have been reported (Lebeau *et al.*, 1998). The advantages associated with this combination, *i.e.* higher permeate quality through combined sorption and biodegradation, higher process stability, lower fouling susceptibility and enhanced removal of microbiological agents, have prompted its emergence as and advanced wastewater reclamation technology (Seo and Ohgaki, 2001; Seo *et al.*, 1996).

4.3. Bottlenecks of membrane processes

Membrane processes have many advantages over conventional tertiary treatment technologies: higher treated water quality, small footprint of the plant, flexibility of operation and lower energy consumption. However, two bottlenecks can be identified:

- As disposal options are being restricted, *treatment of the produced concentrates* is rapidly becoming one of the main factors influencing the overall feasibility of any membrane application (Squire *et al.*, 1996).
- *Fouling* is a major operational concern, leading to substantial technical problems and economic loss. It can be controlled, but needs attention to feed pre-treatment, membrane and module selection and operating strategies (Fane *et al.*, 2000). In this respect, biofouling has been termed “the Achilles heel of membrane processes” (Flemming *et al.*, 1997).

5. EVAPORATION

5.1. Overview

Evaporation is by its broadest definition the conversion of a liquid to a vapour and applies to such widely diverse equipment as boilers, cooling towers, dryers and humidifiers, and losses from fields, storage tanks and reservoirs. In the narrower chemical engineering sense, evaporation is the removal of a volatile solvent from a solution or a relatively dilute slurry by vaporising the solvent (Standiford, 1998). As a water cleansing process, it mimics the natural water reuse in the hydrologic cycle (Buros, 2000).

There are two primary types of evaporators: mechanical evaporators and evaporation ponds. Evaporation ponds do not need a power source, but they do not provide the opportunity for water recovery (Ahmed *et al.*, 2000). As shown in Figure 4, mechanical evaporators require an input of energy in the form of electricity, steam, solar or ambient heat (Chaibi, 2000; Virk *et al.*, 2001).

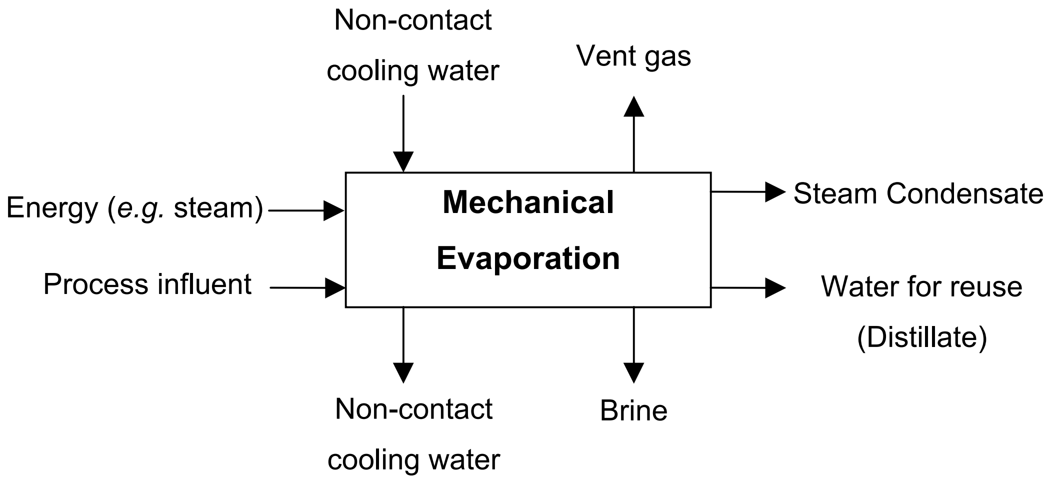

Figure 4 Schematic diagram of a mechanical evaporation process (Zinkus *et al.*, 1998)

Most of the complexity and cost of a mechanical evaporation installation is a result of attempts to reduce energy consumption, which is usually by far the most important element of operating cost. By reusing the vapour as a heat source, energy consumption can be significantly reduced. In practice, this can be achieved by using the inherent heat of the vapour directly in another evaporation chamber (multi-stage flash or multiple-effect evaporation) or by recompressing the vapours formed (Buros, 2000). Through the use of innovative materials in the heat-exchanger and improvements in the process design, the energy consumption can be as low as 6 - 8 kWh/m^3 (Breschi, 2000; Koistinen, 1999; Ramm-Schmidt, 1998; Wade, 2001).

5.2. Evaporation as a wastewater reclamation technology

In wastewater treatment, evaporation is used to reduce the volume of liquid waste streams, such as reverse osmosis concentrates (Baumgarten and Seyfried, 1996; Borsani *et al.*, 1996), landfill leachates (Birchler *et al.*, 1994; Wang, 1998) and pulp and paper mill wastewaters (McKeough and Fagernas, 1999; Ramm-Schmidt, 1998), thus reducing discharge costs.

Some recent applications, however, have seen the implementation of this technology for reuse of domestic wastewaters. For example, Anda *et al.* (2001) described the use of evapotranspiration as a domestic wastewater reclamation technology for remote Australian indigenous communities, while Tleimat and Tleimat (1996) studied the efficiency of a small vapour recompression distillation unit for grey water reclamation. A major study was undertaken by Rose *et al.* (1999), who studied the integration of evaporation in an advanced WWTP effluent reclamation facility. Evaporation, used as an additional sterilisation step, was found to result in inactivation of viruses and bacteria due to the heat and separation of the process.

5.3. Bottlenecks of evaporative processes

Water recovered from an evaporative treatment can be clean enough to be reused or discharged into the environment as all non-volatile substances can be completely separated from the distillate stream. Evaporation is perhaps the one treatment technology that most completely reduces the widest range of drinking water contaminants. In addition, it is a simple, extremely robust and reliable technology. However, disadvantages include:

- Despite developments in evaporation technology, *operational* (*i.e.* energy) *and capital costs* are high. To date, this limits the application of evaporation to large-scale units in arid areas.
- *Foaming, fouling, scaling* are typical operational difficulties that may require auxiliary systems (*e.g.* pre-treatment), which can increase costs.
- Although evaporation is well suited for waters containing inorganic salts and heavy metals, special care must be taken concerning *volatile compounds* such as TAN and VOCs.

6. ELECTROLYSIS

6.1. Overview

Electrochemistry offers promising approaches in the treatment of effluents containing harmful components. The inherent advantage is its environmental compatibility, due to the fact that the main reagent, the electron, is a "clean reagent" and there often is no need for adding extra reagents (Juttner *et al.*, 2000). Other advantages include (Grimm *et al.*, 1998; Rajeshwar *et al.*, 1994):

- *Amenability to automation:* the system variables of electrochemical processes, *i.e.* cell current and electrode potential, are particularly suited for facilitated data acquisition, process automation and control.
- *Energy efficiency*: electrochemical processes generally have lower temperature and pressure requirements than their non-electrochemical counterparts, *e.g.* thermal incineration or supercritical oxidation.
- *Cost effectiveness*: the required equipment and operation are generally simple, and if properly designed, also inexpensive.
- *Versatility:* a wide array of electrochemical techniques exist, including disinfection, cathodic reduction of inorganic constituents and metal cations (mostly heavy metals), direct and indirect anodic oxidation of organic or inorganic contaminants, precipitation, separation through membranes using an electrical field,.... In addition, these processes are applicable to a wide variety of media and pollutants in gases, liquids, and solids and treatment of small to large volumes of microlitres up to millions of litres.

6.2. Anodic oxidation processes

By anodic oxidation of wastewater either a partial reduction of the toxicity (*i.e.* electrochemical conversion) or a complete decomposition of the pollutants (*i.e.* electrochemical incineration to water and CO_2) can be achieved (Comninellis, 1994). The attempts for an electrochemical oxidative treatment can be subdivided into 2 categories: direct oxidation at the anode and indirect oxidation using appropriate electrogenerated oxidants. The schemes of these 2 processes are depicted in Figure 5.

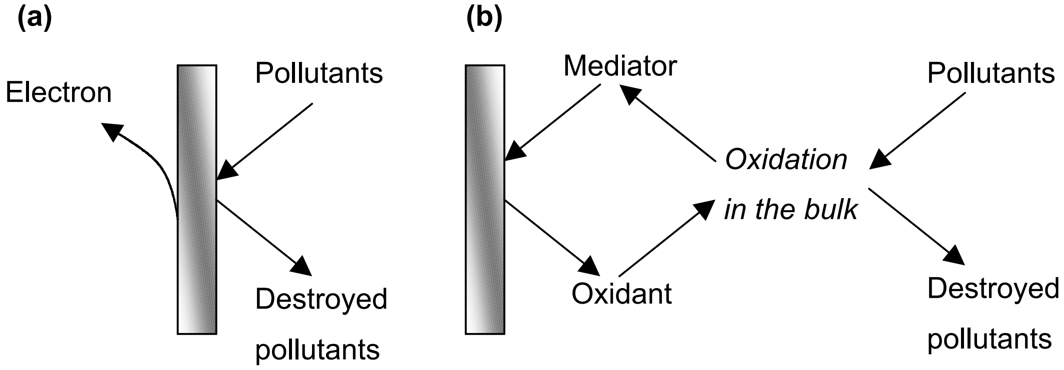

Figure 5 Scheme of pollutant removal through (a) direct anodic oxidation and (b) indirect bulk oxidation (Chiang *et al.*, 1995)

6.2.1. Direct anodic oxidation

A comprehensive model describing the direct oxidation of an organic molecule with simultaneous oxygen evolution allows the distinction between 2 limiting cases: “active” and “non-active” electrodes (Comninellis, 1994; Foti *et al.*, 1999; Simond *et al.*, 1997). A schematic diagram of the model is presented in Figure 6.a.

The initial step (reaction 1) is the discharge of water molecules to form adsorbed hydroxyl radicals. Oxygen evolution can take place either by electrochemical oxidation of physisorbed hydroxyl radicals (non-active pathway, reaction 3) or by electrochemical oxidation of the surface itself followed by the release of the chemisorbed oxygen in a chemical decomposition reaction (active pathway, reactions 2 and 4). The non-active pathway requires the potential of the H_2O/H_2O_2 redox couple, while the active pathway may be available at a lower potential, that of a redox couple M/MO, provided that such a couple exists (*e.g.* M = RuO_2, Pt, IrO_2). Obviously, diamond and fully oxidised metal oxides (*e.g.* M = PbO_2, SnO_2) can only exhibit the non-active pathway. The oxidation of an organic species, R, competes with oxygen evolution and may also occur via 2 alternate ways:

- At non-active electrodes, the oxidation is step mediated by physisorbed (•OH) radicals, essentially yielding fully oxidised products such as CO_2 and H_2O (Comninellis and Pulgarin, 1993). Hence, non-active electrodes favour electro-combustion and may be ideal electrodes for wastewater treatment.
- Active electrodes result in a lower potential because of the formation of the redox couple M/MO, thus limiting the accumulation of (•OH) radicals, hence also preventing the combustion of organics. Indeed, by similarity with the heterogeneous catalytic oxidation with O_2 on an oxide catalyst, selective

oxidation according to reaction 6 is expected (Comninellis and Pulgarin, 1991). Consequently, active electrode surfaces favour electrochemical conversion and may be suited for electrosynthesis.

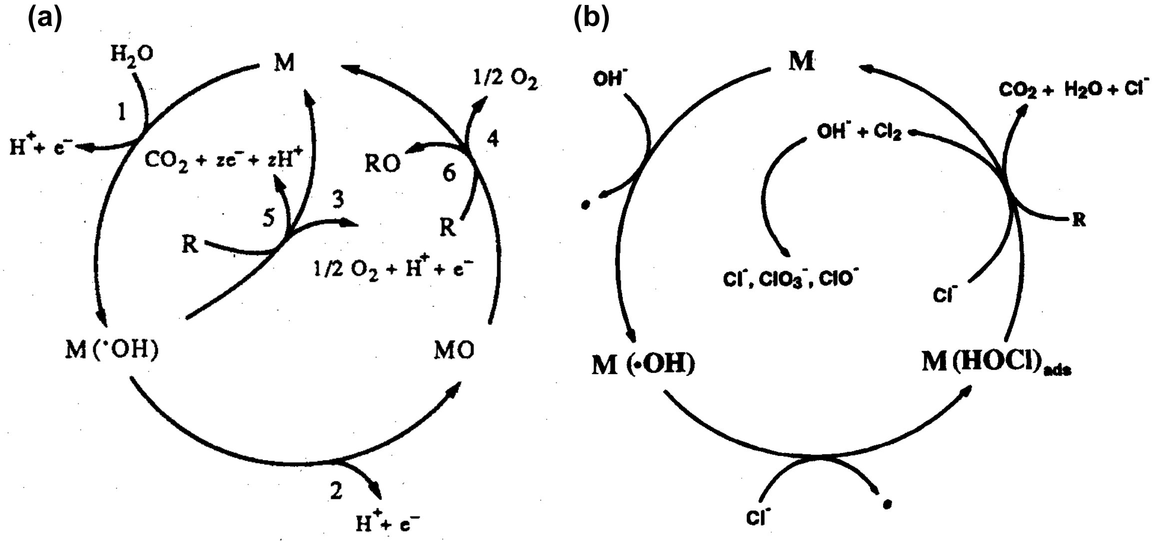

Figure 6 Mechanism of (a) anodic oxidation of organic compounds with simultaneous oxygen evolution (Comninellis, 1994; Foti *et al.*, 1999) and (b) anodic oxidation at non-active electrodes in chloride-containing media (Bonfatti *et al.*, 2000b)

Recently, Bonfatti *et al.* (2000b) proposed an analogous model for the direct anodic oxidation of organics in chloride-containing media with simultaneous chlorine evolution (Figure 6.b). In this model, the oxygen transfer is carried out by adsorbed oxychloro-species instead of hydroxyl radicals.

6.2.2. Indirect bulk oxidation

In an indirect oxidation process, an electrochemically generated oxidant is used to shuttle electrons between the pollutant substrate and the electrode surface (Figure 4.b). The most used oxidant is chlorine or hypochlorite (Chiang *et al.*, 1995), but ozone (Malik *et al.*, 2001), Fenton's reagent (Huang *et al.*, 1999), silver (Panizza *et al.*, 2000b) and peroxodisulphuric acid (Michaud *et al.*, 2000) can also be produced electrochemically. In a chloride-containing medium, anodic oxidation of chloride and subsequent formation of chlorine / hypochlorite is readily observed:

$$2\,Cl^- \rightarrow Cl_2 + 2\,e^- \qquad (1)$$

$$Cl_2 + H_2O \rightarrow OCl^- + Cl^- + 2\,H^+ \qquad (2)$$

OCl^- is a powerful oxidant that can achieve substrate destruction in the aqueous bulk phase. Again, 2 limiting pathways can be identified (Comninellis and Nerini, 1995):

- For active electrodes, the electrogenerated chlorine supplements the selective electroconversion, further oxidising the formed intermediates. Initially, this reaction rate is high, and no OCl^- can be detected. When the rate of bulk hypochlorite oxidation with the more reactive organics has become very low, chlorine is evolved in the electrolyte. It is said that the chlorine demand has been satisfied and in wastewater treatment, this process is known as breakpoint chlorination (Kraft *et al.*, 1999b).
- For non-active electrodes, bulk reaction of organics with electrogenerated OCl^- can also take place, although the direct combustion mechanism according to the scheme of Figures 6.a and 6.b persists.

Consequently, the catalytic effect of the addition of chloride to the electrolyte is more pronounced for active electrodes (Comninellis and Nerini, 1995; Iniesta *et al.*, 2001a; Panizza *et al.*, 2000a).

6.3. Electrochemical treatment of water and wastewater

The electro-oxidation of organic model pollutants has been extensively documented. For example, Comninellis and Pulgarin studied the direct degradation of phenol at non-active SnO_2 electrodes (1993) as well as active Pt electrodes (1991). The catalytic effect of adding chloride or small amounts of H_2O_2 on the phenolic removal efficiency has also been scrutinised (Comninellis and Nerini, 1995; Iniesta *et al.*, 2001a; Lin *et al.*, 1998). Bonfatti *et al.* (1999, 2000b) evaluated different anodes and the role of active chlorine mediation for the electrochemical incineration of glucose.

Many industrial and biorefractory wastewaters have been successfully treated using the electrochemical method. Examples include landfill leachates (Chiang *et al.*, 1995; Wang *et al.*, 2001), textile dye solutions (Shen *et al.*, 2001; Vlyssides *et al.*, 2000), plating rinse effluents (Verhaege *et al.*, 2002), chemical (Panizza *et al.*, 2000a) and olive oil manufacturing wastewaters (Saracco *et al.*, 2001). Because many of these wastewaters invariably contain substantial amounts of chloride, the indirect oxidation effect plays an important role.

Electrochemical reactors have also been used for the removal of inorganic species from wastewater. Nitrite can be electrochemically oxidised to nitrate, while TAN can be removed either by indirect or direct oxidation to N_2 (Kim *et al.*, 2001; Lin and Wu, 1996; Marincic and Leitz, 1978). Other electrochemical wastewater treatment processes rely on the increase in pH caused by the cathodic hydrogen evolution reaction, which can lead to the formation and subsequent precipitation of insoluble salts such as $CaCO_3$, $Mg(OH)_2$ and $MgNH_4PO_4$. Examples include electrochemical sewage treatment for the removal of TAN and phosphate (Dellamonica *et al.*, 1980) and electrochemical drinking water softening (Bannoud *et al.*, 1993).

As an alternative to chlorination, electrochemical disinfection of water and wastewater has received considerable attention (Kraft *et al.*, 1999a,b; Matsunaga *et al.*, 2000; Okochi *et al.*, 1997). The main advantage is the *in situ* generation of disinfective chemicals, which eliminates the need for storage and transport of dangerous chlorine (Patermarakis and Fountoukidis, 1990). The prime reagent is electrogenerated hypochlorite, although electrical discharges can also generate UV radiation, ozone, hydrogen peroxide and other disinfective species (Anpilov *et al.*, 2001). The process can successfully inactivate waterborne pathogens such as *Cryptosporidium* (Venczel *et al.*, 1997) and hepatitis A virus (Morita *et al.*, 2000).

6.4. Emerging electrode materials

The development of suitable electrode materials for electrochemical wastewater treatment opens up a challenging field for materials science. An ideal wastewater electrode is electrically conductive, chemically inert, microstructurally stable and has a high overpotential for oxygen evolution.

Because of its unique properties, diamond offers exciting new possibilities as an electrode material (Rajeshwar *et al.*, 1994). Although diamond is extremely hard, resistant to corrosion, heat and radiation, optically transparent and thermally conductive, its insulating character ($> 10^{12}\ \Omega$cm) would normally preclude its use in electrochemistry. However, by doping with boron, the resistivity of this material can be made as low as $0.01\ \Omega$cm. Consequently, the promising characteristics of boron-doped diamond (BDD) electrodes have been the focus of a body of scientific studies. The research group of Comninellis extensively tested BDD electrodes with respect to the oxidation of 2-naphtol (Panizza *et al.*, 2001a), 4-chlorophenol (Gherardini *et al.*, 2001; Rodrigo *et al.*, 2001), carboxylic acids (Foti *et al.*, 1999; Gandini *et al.*, 2000) and 3-methylpyridine (Iniesta *et al.*, 2001b; Panizza *et al.*, 2001b). In addition, the preparation of electro-oxidants such as silver (Panizza *et al.*, 2000b), peroxodisulphuric acid

(Michaud *et al.*, 2000) and chlorine (Ferro *et al.*, 2000) was studied. These reports confirm the potential of BDD as wastewater treatment electrode:

- Although some oxygen-containing functional groups can be formed on virgin diamond, the electrode is stable in a wide variety of electrolytes.
- Direct electrochemical incineration initially proceeds at high energetic efficiency; at lower organics concentrations, the current efficiency decreases because of mass transport limitations.
- BDD electrodes are very efficient chlorine producers.
- Because of its weak adsorptive properties, the hydroxyl or oxy-chloro radicals formed on BDD electrodes are very reactive towards the oxidation of organics.

6.5. Bottlenecks of electrolysis

In the electrochemical abatement of wastewater, two factors are of major concern:

- The *energy requirements* of electrochemical processes can be high, mainly due to parasitic side reactions and ohmic losses. Especially relatively dilute electrolytes necessitate the application of high cell potentials. The addition of salts (*e.g.* NaCl) efficiently reduces the energy demand and catalyses the indirect oxidative abatement, but may render the electrolyte inept for subsequent discharge into the environment (Saracco *et al.*, 2001).
- The possible formation of *organochlorinated* and *DBP-like products* is a critical issue. Although some studies argue that these components are further oxidised to unharmful constituents (Bonfatti *et al.*, 2000b; Comninellis and Nerini, 1995) this point merits further research.

CHAPTER 4

PRODUCTION OF HIGH QUALITY RECLAIMED WASTEWATER THROUGH EVAPORATIVE TREATMENT[1]

ABSTRACT

The present study explored the application of evaporative technology as an alternative desalination technique for wastewater treatment plant effluent. In the experiments carried out, it was shown that evaporation allowed for extensive decontamination of wastewater treatment plant effluent as most inorganic and organic contaminants were completely removed. TAN could be retained in the concentrate when pH was adjusted to values lower than 4; however this led to increased nitrite volatility due to the formation of nitric oxide. Using an electronic nose, it was observed that evaporation gave rise to a change in odour quality and quantity due to volatilisation of the volatile organic compounds present in the effluent. Remediation of these components was carried out by Fenton oxidation prior to evaporation. A high removal was observed under the applied conditions of Fe^{2+} = 25 mg/L, H_2O_2 = 50 mg/L, pH = 3 and a reaction time of 2 hours.

Keywords: evaporation, WWTP, effluent, electronic nose, Fenton

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[1] Redrafted after Van Hege K., Dewettinck T. and Verstraete W. (2001) Pre-evaporative Fenton remediation of treated municipal wastewater for reuse purposes. *Environmental Technology*, **22** (5), 523-532.

INTRODUCTION

The continuously growing demand for clean water, more stringent quality requirements and the decreasing availability of suitable natural water resources are prompting industry and government to explore new innovative ways of water conservation. An approach that has received considerable attention in recent years, is the reuse of treated effluent from wastewater treatment plants (WWTPs) in process water (Filteau *et al.*, 1995) and groundwater recharge applications (Crook *et al.*, 1999; Van Houtte *et al.*, 1998).

Treatment of WWTP effluent to reclaimed water requires partial or total removal of the organic, inorganic, suspended and biological pollutants present. Nowadays, a variety of advanced biological, physical and chemical technologies are available to produce reclaimed water of any desired quality (Weber and LeBoeuf, 1999; Zinkus *et al.*, 1998). The simplest systems involve solid/liquid separation processes and disinfection, whereas more complex reclamation systems involve combinations of physical, chemical, and biological processes employing multiple barrier treatment approaches for contaminant removal (Asano and Levine, 1995; Vandevivere *et al.*, 1998). In recent years, many researchers have investigated the application of membrane technology in wastewater reuse (Johnson *et al.*, 1997; Visvanathan *et al.*, 2000). The filtrative treatment of WWTP effluent by microfiltration (Ahn and Song, 1999), ultrafiltration (Tchobanoglous *et al.*, 1998; van Hoof *et al.*, 1998), reverse osmosis (Abdel-Jawad *et al.*, 1999) as well as the *in situ* combination of membrane filtration and activated sludge processes (Cicek *et al.*, 1998; Côté *et al.*, 1997) have been extensively documented. It has been shown that an efficient combination of these techniques is suitable for effluent desalination, resulting in the production of high quality reclaimed water (van Hoof *et al.*, 1999; Van Houtte *et al.*, 1998). Recently, Rose *et al.* (1999) reported the use of evaporation as an alternative effluent desalination technique.

In wastewater treatment, evaporation is often used to reduce the volume of liquid waste streams, such as landfill leachates (Birchler *et al.*, 1994), reverse osmosis (Baumgarten and Seyfried, 1996) or process-water (McKeough and Fagernas, 1999) concentrates. Recent advances in the design and efficiency of evaporators have resulted in the implementation of this technology for reuse of wastewater and treatment of dilute wastewater streams. Evaporation is well suited for waters containing heavy metals, inorganic salts and suspended solids. Yet special care has to be taken when treating wastes containing organic contaminants, especially volatile organic compounds (VOCs) (Zinkus *et al.*, 1998).

VOCs, originating from industrial, commercial, public and residential discharges to municipal wastewater collection systems, are an important group of contaminants present in WWTP effluent. Using GC-MS, Paxeus (1996) detected and identified 137 VOCs in the effluent of 3 large WWTPs in Sweden. However, the duration and complexity of GC-MS analyses has prompted researchers to develop and explore simple and rapid sensors. In recent publications describing the use of an electronic nose for measurement of VOCs in wastewater, Stuetz *et al.* (1999a,b) and Dewettinck *et al.* (2001b) were able to correlate electronic nose analyses to various water quality parameters.

Possible remediation techniques for VOCs include adsorption on granular activated carbon (GAC), air stripping and advanced oxidation processes (AOP), such as ozonation or Fenton's reagent. The latter process, which consists of the generation of highly reactive hydroxyl radicals through the iron-catalysed decomposition of hydrogen peroxide according to equations (1) and (2) (Fenton, 1894), has been applied to a variety of toxic and refractory organic components.

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + HO\bullet \tag{1}$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + H^+ + HO_2\bullet \tag{2}$$

The observed effects are: organic pollutant destruction, toxicity reduction, disinfection, biodegradability improvement, BOD/COD removal and odour and colour removal. Lin *et al.* (1999) found that Fenton's reagent was able to remove surfactants present in wastewater, while Huston and Pignatello (1999) observed complete degradation of selected pesticide active ingredients such as atrazine, captan, alachlor and methoxychlor. The toxicity reduction following Fenton treatment was evidenced by Kochany and Lugowski (1998), who examined the removal of nitrification inhibitors from polluted groundwater. Buyuksonmez *et al.* (1998) modelled the disinfecting capacity of modified Fenton reactions on *Xanthobacter flavus* FB71. Finally, Park *et al.* (1999) demonstrated a drastic increase of the biodegradability of a Fenton remediated pigment wastewater.

It was the aim of this study to investigate the application of evaporation as an advanced treatment technique for the direct production of high quality reclaimed water. Furthermore, the remediation of VOCs present in WWTP effluent by Fenton's reagent was evaluated using an electronic nose.

MATERIALS AND METHODS

WWTP effluent and reagents

WWTP effluents were obtained from the municipal WWTPs of Ghent and Deurne (Belgium). Sewage treatment in both plants consists of primary sedimentation, followed by conventional activated sludge treatment and gravitational clarification. To exclude possible changes in composition, samples were stored at 4 °C and processed within 5 days. Sulphuric acid, $FeSO_4.7H_2O$ (VEL, Belgium) of analytical grade (> 98 % w/w) and H_2O_2 (30 % w/w; VEL, Belgium) were used in the experiments.

Evaporation experiments and related parameters

One-day experiments were carried out on 2 WWTP effluents using an industrial evaporator/concentrator (LED ITALIA R 150, Pordenone, Italia) with a capacity of 0.17 m³/day. During each experiment, a flow proportional pooled sample (5 sub-samples) was taken of the WWTP effluent and the evaporate. The evaporator operated at 35 °C and a residual pressure of approximately 5.3 kPa.

Additional lab-scale batch experiments were performed using a Rotavapor (Büchi, Flawil, Switzerland) operated at 100 °C. When using a batch evaporation device, the concentration factor (CF) was defined as the ratio of the original effluent volume ($V_{effluent}$) to the volume of the concentrate after evaporation ($V_{concentrate}$):

$$CF = \frac{V_{effluent}}{V_{concentrate}} = 1 + \frac{V_{evaporate}}{V_{concentrate}} \tag{3}$$

Since $V_{effluent} = V_{concentrate} + V_{evaporate}$. The percentage removal was calculated as the ratio of the amount retained in the concentrate to amount originally present in the effluent:

$$\%_{removal} = \frac{1}{CF}\frac{C_{concentrate}}{C_{effluent}} * 100 = \left(1 - \frac{CF - 1}{CF}\frac{C_{evaporate}}{C_{effluent}}\right) * 100 \tag{4}$$

Because $V_{effluent}*C_{effluent} = V_{concentrate}*C_{concentrate} + V_{evaporate}*C_{evaporate}$, with C the concentration. Batch experiments consisted out of evaporation of 0.5 L effluent and unless stated otherwise, a concentration factor of 2 was applied. For the lab-scale batch experiments, samples were taken of the WWTP effluent, evaporate and concentrate. Lab-scale experiments were carried out at atmospheric pressure and 100 °C.

General parameters

The WWTP effluents as well as the evaporates and concentrates were analysed for a number of routine parameters. Nitrite, nitrate, total ammonia nitrogen (TAN), chloride, phosphate, sulphate, total organic carbon (TOC), chemical oxygen demand (COD), suspended solids (SS), pH, conductivity and turbidity were determined according to the Standard Methods for the examination of water and wastewater (AHPA, 1992). For some samples, the oxidisability was determined using potassium permanganate instead of potassium dichromate, as this allows for a more sensitive COD determination with a lower detection limit (0.8 vs. 5 mg O_2/L).

Plate counts were performed using the spread plate technique (AHPA, 1992). The following agars, incubation times and temperatures were used: Nutrient agar (Oxoid, England), 3 days at 37 °C for the total count; Mc Conkey agar (Difco, USA), 1 day at 43 °C for *E. coli*; Slanetz & Bartley agar (Oxoid, England), 1 day at 37 °C for faecal streptococci.

Presence of NO / NO_2

Nitric oxide was measured using Gastec 1010 nitric oxide detection tubes (Scantec, Belgium). In this tube, nitric oxide (NO) is oxidised by chromic acid to nitrogen dioxide (NO_2) which oxidises o-tolidine to nitroso-o-tolidine (yellow product).

Electronic nose analyses

On the WWTP effluents and evaporates, electronic nose measurements as described by Dewettinck *et al.* (2001b) and Maricou *et al.* (1998) were carried out. The electronic nose (FOX 3000, Alpha M.O.S., Toulouse, France) contains 12 metal oxide sensors, each of which displays a good sensitivity towards a broad range of volatile compounds. The sensors used as well as the group of compounds to which they are sensitive, are listed in Table 1.The measurement principle of an electronic nose is based on the change in electrical resistance of the sensors when volatile compounds are present.

The metal oxide sensors are semiconductors and are gas sensitive: oxygen in the air is chemisorbed on vacancies in the lattice of the bulk material and removes electrons from the conducting band (eq. (5)):

$$\text{sensor electron} + 1/2\ O_2 \rightarrow O^- \text{ (s)} \qquad (5)$$

In the presence of a gas or a fragrant molecule (G), this chemisorbed oxygen (O^-) reacts irreversibly to produce combined molecules (GO) (eq. (6)):

$$G(g) + O^- \text{ (s)} \rightarrow GO \text{ (g)} + \text{sensor electron} \qquad (6)$$

The liberated electrons reduce the potential barrier of the oxide grains, which leads to a decrease in resistance. The size of the response depends on the nature of the detected molecules, their concentration and the type of sensor. The response time depends on the reaction kinetics, the volume of headspace measured and the flow rate of the gas. The flow rate was set at 150 mL/min and the injection time at 60 s.

Table 1 Overview of the sensors in the electronic nose, together with the chemical compounds to which they are sensitive (Alpha M.O.S., 1995)

Sensor	Detection of
P30/1	Polar compounds
P10/1	Hydrocarbons
P10/2	Methane, propane and aliphatic nonpolar molecules
P40/1	Chlorinated and fluorinated compounds
P40/2	Aldehydes
PA3	Hydrogen bounds, smoke detection
P70/0	Food aromas, alcohol and aromatic compounds
T50/3	Ammonia derivatives and amine containing compounds
PA2	Low concentrations of hydrogen, H_2S and ammonia
T50/1	S-containing compounds
T40/1	Chlorinated and fluorinated compounds
T70/2	Alcohol vapours and aromatic compounds

Samples (200 mL in a 500 mL borosilicated glass bottle) are heated to 90 °C to promote volatilisation of the VOCs. Prior to measurement of the headspace of a sample, a carrier gas (i.e. synthetic air) is sent over the sensors until the baseline, corresponding to an initial resistance R_0, is reached. Upon injection of the gaseous headspace of the sample, the resistance of the sensor drops to a minimal value R. For each sensor, the sensorial odour perception (SOP, eq. (7)) is expressed as the maximal proportional change in resistance with regard to the base line resistance:

$$\text{SOP}\ (\%) = 100 * \frac{R_0 - R}{R_0} \tag{7}$$

The total odour quantity of the sample is determined by the reaction on all 12 sensors and is expressed as SOP_{av} (eq (8)).

$$\text{SOP}_{av} = \frac{1}{V_{gas}} \frac{1}{12} \sum_{i=1}^{12} \text{SOP}_i \tag{8}$$

Where SOP_i is the SOP value of sensor i (%) and V_{gas} the volume of headspace gas injected (in this study 0.150 L). The quality of the odour is visualised in radar plots, in which the SOP of each sensor is plotted in a figure with 12 axes, one for each sensor. As each sensor is sensitive towards a certain group of volatile compounds, the form of the radar plot is an indication for the nature of the volatile compounds, a so-called fingerprint. For each experiment, demineralised water was measured as a reference.

RESULTS

Experiments with an industrial pilot-scale evaporator

The results of the one-day experiments carried out on the effluents of Deurne and Ghent are reported in Table 2. For both effluents, the removal of most inorganic species is complete: contaminants such as sulphate, chloride and nitrate are removed to values below the detection limit of the methods used. Nitrite removal is high (83 and 99 % for Ghent and Deurne evaporates, respectively) and only trace amounts remain in the evaporate. No phosphate or heavy metals (Pb, Mn, Fe, Zn, Hg) could be detected in the effluents or evaporates (results not shown). However, no TAN removal is achieved. The high degree of demineralisation is reflected in the measured electrical conductivity, which decreases by 99 and 81 % for the Ghent and Deurne effluent, respectively. The higher residual conductivity of the Deurne evaporate is caused by the presence of a significant amount of TAN, which also seems to have an effect on the observed pH, as a lower pH-decrease is noted.

Complete suspended solids removal is achieved and COD and TOC of the evaporates are below the respective detection limits. In both cases, a water of high clarity is produced, since the turbidity is lower than 1 NTU and no significant absorption is observed in the colour scans (results not shown). No faecal indicator organisms are detected, yet a low total heterotrophic count, corresponding to a removal of 99.60 and 99.96 % for the Ghent and Deurne evaporates respectively, is observed.

Table 2 Results of the pilot scale experiments carried out on 2 WWTP effluents. X = not determined

		Ghent		Deurne	
		Effluent	**Evaporate**	**Effluent**	**Evaporate**
SO_4^{2-}-S	mg/L	52	<3	57	X
Cl^-	mg/L	93.4	< 3	118	X
TAN	mg N/L	0.238	0.247	25.4	27.5
NO_3^--N	mg/L	3.94	<0.23	1.13	<0.23
NO_2^--N	mg/L	0.091	0.015	0.323	0.018
pH	/	7.82	5.35	7.82	7.32
EC	µS/cm	936	12	1215	233
TOC	mg C/L	4.2	<2	24	<2
COD_{total}	mg O_2/L	21	<5	67	<5
SS	mg/L	15	<5.2	55	<5.2
Turbidity	NTU	1.9	0.6	11.1	0.2
Total count	CFU/mL	$2.8 *10^4$	$1.1 *10^2$	$1.0 *10^6$	$4.2 *10^2$
E. Coli	CFU/mL	$3.3 *10^2$	<1	$1.9 *10^4$	<1
Faecal streptococci	CFU/mL	$8.0 *10^1$	<1	$2.7 *10^3$	<1

Lab-scale experiments

Influence of pH

Batch evaporation experiments were carried out at different pH-values. Prior to evaporation, the pH of the WWTP effluent was set at a specific value using sulphuric acid. The influence of pH on the volatility of different compounds is shown in Table 3. pH has no significant influence on COD removal, as almost all the COD is retained in the concentrate and evaporate COD values average around 2.2 mg O_2/L. TAN is only retained in the concentrate when pH is low (96 % removal at pH 3 compared to 0 % at pH 7). Low pH values, however, lead to an increased volatility of nitrite (55 % removal at pH 3 compared to 95 % at pH 7). During the experiment at pH 3, the presence of nitrous oxide in the gaseous atmosphere was qualitatively detected using a Gastec NO-NO_2 detection tube placed in the evaporation chamber. The tube coloured yellow, indicating the presence of NO (or NO_2).

Table 3 Influence of pH on the removal of contaminants present in WWTP effluent. Ghent WWTP effluent : pH = 7.34; COD = 33 mg O_2 /L; SS = 8.0 mg/L; TAN = 2.29 mg/L; NO_2^--N = 0.193 mg/L; NO_3^--N = 4.34 mg/L. Experiments with CF = 2 and pH of the WWTP effluent set at 7, 5 and 3

			pH		
			7	**5**	**3**
Evaporate	pH	/	6.2	6.45	5.91
	TAN	mg N/L	4.86	4.85	0.156
	NO_3^--N	mg/L	0.405	0.307	0.271
	NO_2^--N	mg/L	0.034	0.027	0.173
	COD	mg O_2/L	2.72	2.32	2.24
Concentrate	pH	/	9.04	8.88	4.85
	TAN	mg N/L	0.092	0.055	4.18
	SS	mg/L	8.0	9.1	10.8
	COD	mg O_2/L	63	62	61

Influence of concentration factor (CF)

The influence of the concentration factor was evaluated and the results are shown in Table 4. To limit the volatilisation of TAN, pH was set to 3 prior to evaporation. Applying a higher concentration factor has no effect on the demineralisation of the evaporate as the removal efficiencies for nitrate, nitrite and TAN are largely unaffected: around 95 % of the TAN and nitrate are removed, while only 35 % of the nitrite is eliminated. Analysis of the concentrate for CF = 10 confirmed that under the applied conditions practically all the TAN and nitrate are retained, yet most of the nitrite is volatilised (TAN = 19.4 mg/L, NO_3^- -N = 38.0 mg/L, NO_2^- -N = 0.043 mg/L).

Organics removal is high and consistent as the COD of the evaporate is below the detection limit for the different concentration factors applied; moreover, using the permanganate method, the COD of the evaporate was 1.76 mg O_2/L for a CF = 10.

Table 4 Influence of the CF on the removal of contaminants present in WWTP effluent. Ghent WWTP effluent as in Table 3. Experiments with pH set at 3 and CF = 2, 5 and 10.* = permanganate method used

			CF		
			2	**5**	**10**
Evaporate	pH	/	4.47	4.87	4.43
	TAN	mg N/L	0.060	0.083	0.131
	NO_3^--N	mg/L	0.163	0.236	0.224
	NO_2^--N	mg/L	0.247	0.159	0.138
	COD	mg O_2/L	< 5	< 5	1.76 *

Fate of VOCs

To elucidate the fate of VOCs during evaporation, effluent samples and their respective evaporates (CF = 2) were subjected to electronic nose analyses. Figure 1 shows the electronic nose radar plots of Ghent WWTP effluent and its evaporate. The form of the evaporate radar plot differs from the WWTP effluent, indicating a change in odour quality. The increase in odour quantity of the untreated evaporate is reflected in the surface of the radar plots as well as the computed SOP_{av} values: 113 L^{-1} for the WWTP effluent versus 204 L^{-1} for the evaporate. In comparison with the WWTP effluent, the SOP value of a number of sensors more than doubles for the evaporate sample (*i.e.* T50/3, T70/2, P10/2 and P40/2), while other sensors show little change in SOP value (*i.e.* T40/1, P40/1, PA3 and T70/0).

Fenton pre-treatment of VOCs

Preliminary experiments on WWTP effluent using Fenton's reagent showed that more than 88 % COD-removal was achieved under the conditions of pH = 3, H_2O_2 = 25 - 50 mg/L, Fe^{2+} = 25 mg/L and a reaction time set at 2 hours (results not shown). Taking into account the non-selective feature of the hydroxyl radical, it was hypothesised that a similar decrease in odour could be expected. To investigate this hypothesis, two sets of experiments were carried out: after a Fenton pre-treatment of 2 hours (pH = 3, Fe^{2+} = 25 mg/L and H_2O_2 = 25 and 50 mg/L), samples were subjected to evaporation with CF = 2 and the odour profiles of the evaporates were measured using the electronic nose.

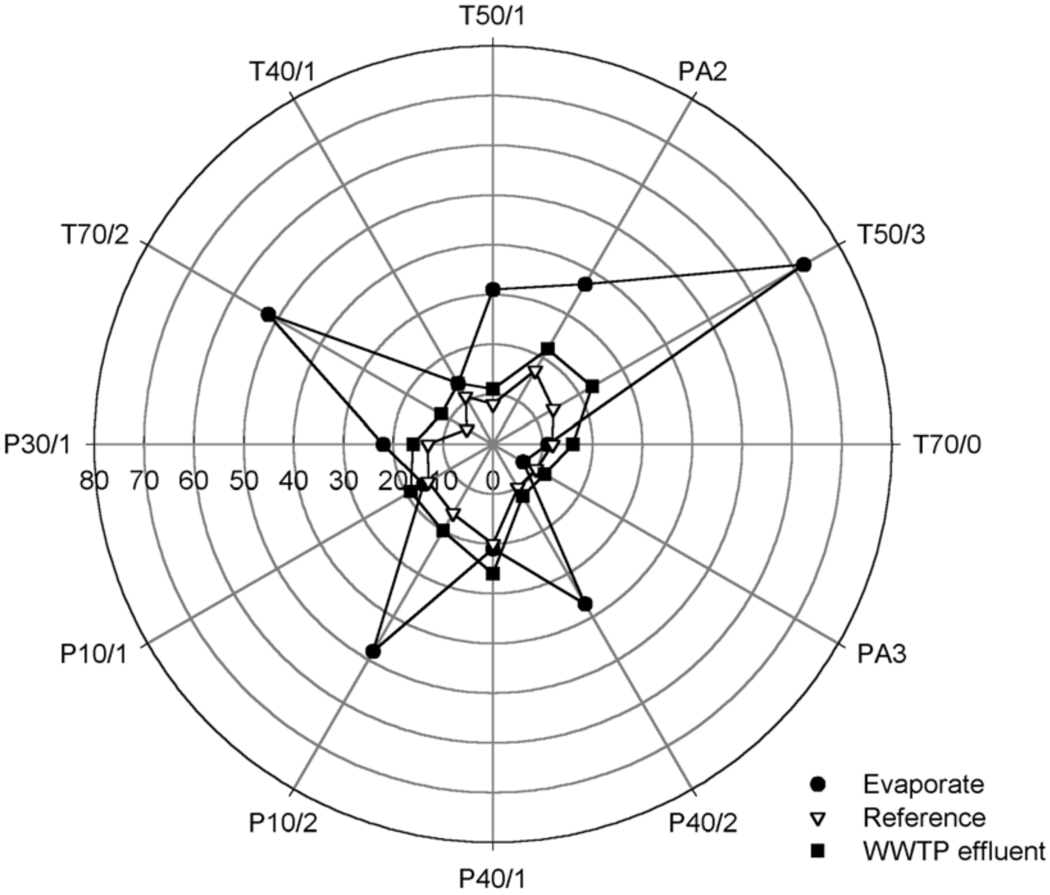

Figure 1 Radar plot of the odour profile of Ghent WWTP effluent, evaporate and reference (demineralised water). Ghent WWTP effluent : pH = 7.63; COD = 53 mg O_2/L SS = 48 mg/L; TAN = 1.32 mg/L; NO_2^--N = 0.068 mg/L; NO_3^--N = 1.92 mg/L. The 12 axes correspond to the 12 different sensors of the FOX 3000 electronic nose. The data points are the relative change in electrical resistance (SOP) of each sensor. Evaporation experiments carried out at pH = 3 and CF = 2

The influence of different H_2O_2-concentrations on the removal of VOCs after the combined treatment (Fenton + evaporation) is shown Figure 2 and Table 5. Compared to the untreated evaporate, a change in odour pattern as well as odour quantity was observed. Applying a H_2O_2-dose of 50 mg/L (pre-treated evaporate 2) decreased the SOP_{av} of the evaporate by 63 %. The measured odour quantity is lower than the non-treated WWTP effluent and comparable to the reference sample (demineralised water) as indicated in Table 5. In addition, the pH correction used for the Fenton pre-treatment resulted in a TAN removal of more than 96 %.

Table 5 Results of the FOX measurements and TAN determinations carried out on untreated and Fenton pre-treated WWTP effluent and the respective evaporates. Ghent WWTP effluent as in Figure 1. Evaporation experiments carried out at pH = 3 and CF = 2. X = not determined

Sample	TAN (mg/L)	SOP$_{av}$ (L^{-1})
Reference (demineralised water)	X	84
Ghent WWTP effluent (untreated)	1.32	113
Evaporate (no pre-treatment)	0.091	204
Pre-treated evaporate 1 (H_2O_2 = 25 mg/L)	0.033	113
Pre-treated evaporate 2 (H_2O_2 = 50 mg/L)	0.068	75

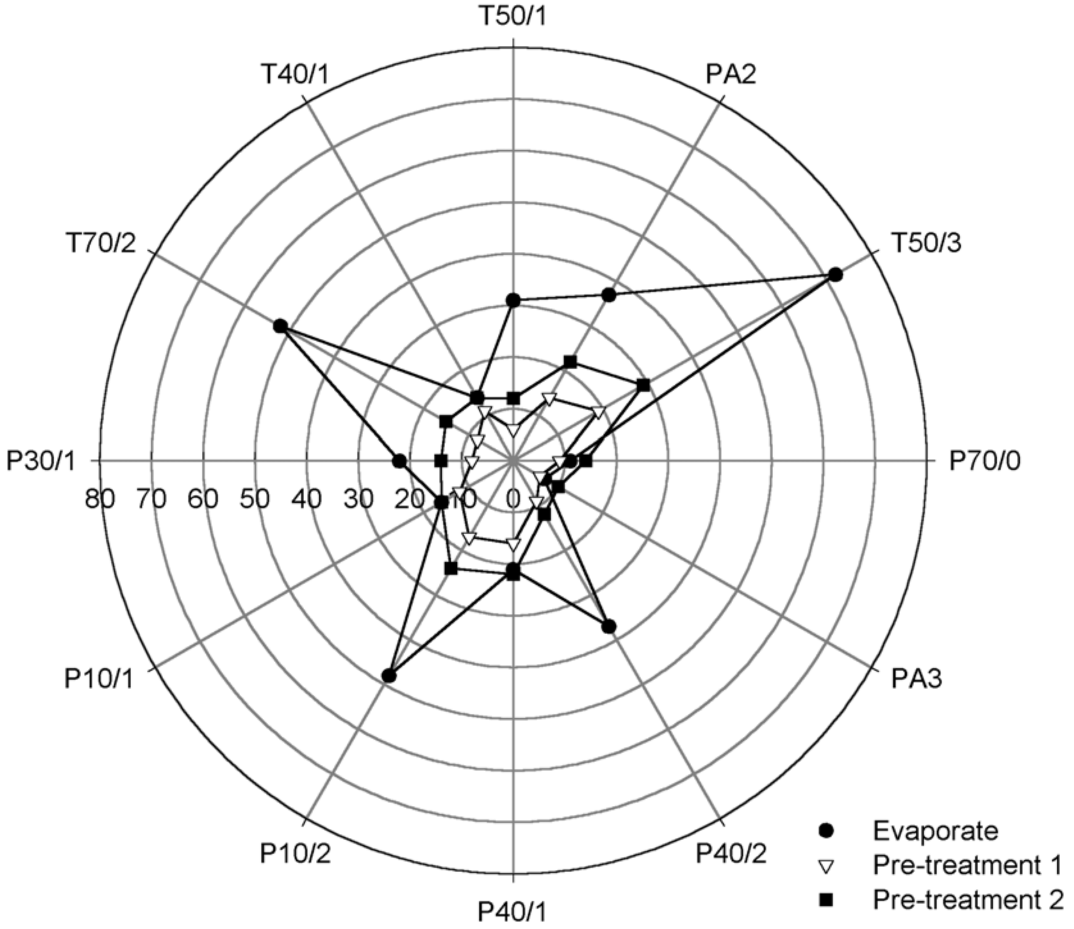

Figure 2 Radar plot of the odour profile of untreated evaporate, pre-treated evaporate 1 and 2. Reference (demineralised water) omitted because of overlapping with pre-treated evaporate 2. WWTP effluent as in Figure 1. The 12 axes correspond to the 12 different sensors of the FOX 3000 electronic nose. The data points are the relative change in electrical resistance (SOP) of each sensor. Evaporation experiments carried out at pH = 3 and CF = 2

DISCUSSION

The production of high-quality reclaimed water from wastewater necessitates the implementation of a combination of advanced treatment technologies. For example, Johnson *et al.* (1997) described the treatment of raw sewage to high quality reclaimed water by a combination of a 200 µm pre-screen, microfiltration and reverse osmosis (RO). Abdel-Jawad *et al.* (1999) reported the reclamation of WWTP effluent using flocculation-sedimentation, sand filtration and reverse osmosis while Van Houtte *et al.* (1998, 1999) employed microfiltration prior to RO treatment. The quality of these RO permeates is similar to that of the evaporates from the continuous tests, showing a very high degree of demineralisation and only traces of residual organic pollution. For example, the decrease in electrical conductivity of 98 % in the study of Van Houtte (1999) is very similar to the values obtained in the present study. Evaporation allows equal or even higher organics removal than RO (permeate COD of 5.3, 13.47 and < 5 mg O_2/L as reported in the respective papers, compared to < 5 in the present study). However, under the conditions of the pilot-scale experiment, no TAN removal was achieved as the TAN present in both effluents is volatilised as ammonia and subsequently condensed in the evaporate according to equation (9) and (10):

$$NH_4^+ + OH^- \leftrightarrow NH_{3,\,aq} + H_2O \qquad pK_a = 9.25 \qquad (9)$$

$$NH_{3,\,aq} \leftrightarrow NH_{3,\,g} \qquad k_H^{25\,°C} = 60\ \text{M/atm} \qquad (10)$$

Where k_H is the Henry's law constant, defined as the ratio of the concentration of a species in the aqueous phase to the partial pressure of that species in the gaseous phase (Sander, 1999).

During the pilot trials, microbiological constituents were also monitored. The disinfective capacities of evaporation have been described previously: in their evaporator pilot study, Rose *et al.* (1999) observed a 99.5 % removal efficiency for the thermoresistant bacteria *Bacillus sterothermophilis* and a 99.9995 % removal of the bacteriophage MS-2, demonstrating extensive inactivation due to heat and separation through the evaporative technology. In the present study, bacterial activity in both evaporates was observed, yet the absence of typical faecal indicator organisms indicates that the observed total count was probably due to bacterial post-treatment contamination in the distillate receiving tank.

In subsequent batch experiments, the influence of pH and CF was further investigated. The observed pH-effect on the volatility of TAN has been reported in literature: Birchler *et al.* (1994) carried out evaporation experiments on different landfill leachates and ammonium acetate solutions and concluded that a 95 – 99 % removal of ammonia could be achieved when pH was set to 4 or lower. While the removal of most inorganic, organic and microbiological contaminants has been studied in the context of wastewater evaporation, to our knowledge, no report has been made concerning the fate of nitrite. At lower pH-values, the nitrite/nitrous acid equilibrium (eq. (11)) shifts towards nitrous acid, which decomposes - especially at elevated temperatures - according to eq. (12) (Thiemann *et al.*, 1987):

$$HNO_2 \leftrightarrow NO_2^- + H^+ \qquad pK_a = 3.34 \tag{11}$$

$$3\ HNO_2 \leftrightarrow HNO_3 + 2\ NO + H_2O \tag{12}$$

The nitric oxide can dissolve in the condensed water to form NO_2^-. In the experimental study, the presence of nitric oxide in the gaseous atmosphere was qualitatively confirmed by a Gastec NO-NO_2 detection tube placed in the evaporation chamber. In Figure 3, a visual representation of the equations (9) and (11) at 25 °C is given. Theoretically, neutral pH values should allow the retention of both TAN and nitrite in the concentrate. However, the temperature increase during evaporation has 2 pronounced effects. First, at elevated temperatures, the equilibrium of eq. (9) is shifted towards lower pH values, resulting in a higher amount of aqueous NH_3 for a given pH value (Figure 3). Second, the k_H-value of NH_3 decreases with increasing temperature, leading to an increased gaseous NH_3 concentration according to eq. (10) (Sander, 1999; Postma, 1982). When combined, both effects result in a pronounced increase of the volatility of ammonia during the evaporative treatment. As a consequence, a pH inhibiting the evaporative volatilisation of TAN (by shifting the equilibrium completely towards ammonium) inevitably leads to an increased chance of nitrite volatilisation, and vice versa.

Moreover, the experimental observations indicate that the pH is the main factor determining the volatility of nitrite and TAN as the applied CF does not have a noticeable influence: all the non-volatile components remain in the concentrate, irrespective of the applied CF. Consequently, a CF of 10 – as is often applied in practice (Birchler *et al.*, 1994) - would not adversely effect the quality of the evaporate.

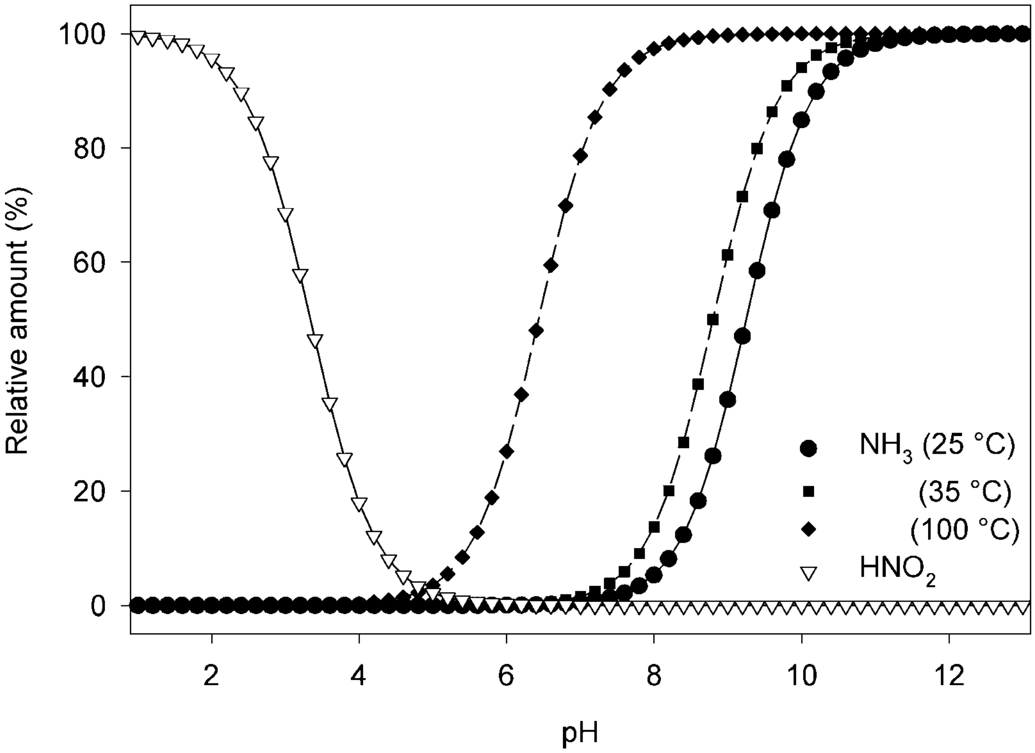

Figure 3 Relative amount of aqueous NH_3 at 25, 35 and 100 °C and HNO_2 at 25 °C as a function of pH according to equations (9) and (11)

The fate of VOCs during evaporative treatment of WWTP effluent was evaluated using an electronic nose. Although the observed increase in evaporate odour quantity could be partly attributed to the applied concentration factor (CF = 2), the change in odour quality is a combination of different effects, in particular desorption and organic decomposition. Because of the nonpolar or moderately polar nature of most VOCs, adsorption of these compounds onto the suspended solids (SS) can be expected (Dewettinck *et al.*, 2001b; Schwarzenbach *et al.*, 1993). As a consequence, desorption can occur due to the stripping effect of the water vapour formed during evaporation. Secondly, it is plausible that part of the (volatile) organic matter subjected to the evaporative thermal treatment decomposes, resulting in the formation of other volatile components (Wu *et al.*, 2001).

From this, it is evident that an evaporation-based treatment of WWTP effluent necessitates the removal of VOCs by a suitable post- or pre-evaporative treatment. In the present study, Fenton's reagent was found to be highly efficient as a pre-evaporative treatment. The observed optimal pH of 3 for Fenton's reagent corroborates with other studies, as most authors report pH values in the range 2 to 4 (Huston and Pignatello, 1999; Kochany and Lugowski, 1998; Lin *et al.*, 1999; Potter and Roth, 1993). Higher pH values lead to precipitation of the formed Fe^{3+} as oxyhydroxides, thus removing the iron from the hydroxyl generating reaction process (Huston and Pignatello, 1999). Reported values of the applied COD/H_2O_2 ratio are in the range of 0.4 - 1.4 (Lin *et al.* (1999) and Kochany and Lugowski (1998), respectively), depending on the extent of detoxification desired. In the present study a COD/H_2O_2 ratio of 1.1 was applied, resulting in a 60 % decrease of odour in the corresponding evaporate. The ratio Fe^{2+}/H_2O_2 can vary between 0.08 (Park *et al.*, 1999) and 0.7 (Lin *et al.*, 1999) and is usually higher for less polluted wastewaters, since a threshold amount of iron catalyst is needed to allow the process to proceed within a reasonable period of time. The WWTP effluent used required a minimal concentration of 25 mg Fe^{2+}/L and a Fe^{2+}/H_2O_2 ratio of 0.5 to allow the reaction to complete within 2 hours.

CONCLUSIONS

It was shown that evaporation guarantees an extensive decontamination of the WWTP effluents under scrutiny, while it is known that it is a simple, robust and very reliable process. Removal of most organic and inorganic contaminants occurs irrespective of the pH and CF applied. The removal of TAN, however, implies setting the pH at 4 or lower and under these conditions nitrite is volatilised. As the presence of VOCs in the evaporate necessitated additional treatment, the removal was evaluated by using a Fenton pre-treatment step. Under the operating conditions of the Fenton oxidation (50 mg/L H_2O_2, 25 mg/L Fe^{2+}, 2 hours of treatment time and an initial pH $\approx$ 3) the resulting evaporate is essentially odour-free. Moreover, the low pH inhibits ammonia volatilisation. The combined process, as schematised in Figure 4, allows for the direct production of high quality reclaimed water from WWTP effluent.

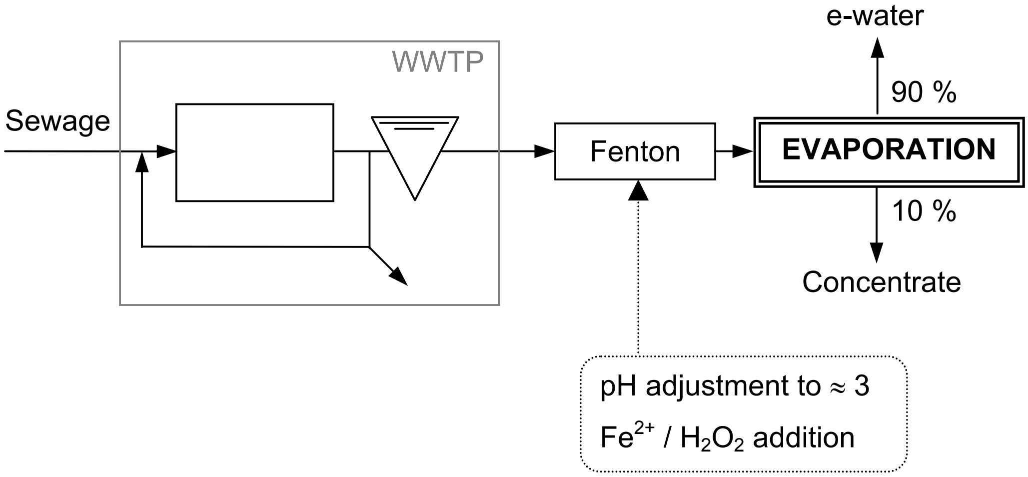

Figure 4 Proposed process scheme

Depending on the price of electricity and/or the availability of steam, predicted costs of planned full-scale evaporators can be as low as 0.17 EUR/m³ evaporate (Al-Shammiri and Safar, 1999), which could make the process competitive compared to filtrative desalination techniques.

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CHAPTER 5

ADVANCED EFFLUENT POLISHING THROUGH FILTRATIVE TREATMENT[1]

ABSTRACT

Membrane processes are increasingly used as an advanced treatment technique for the reclamation of treated domestic wastewater. Despite their inherent advantages, fouling remains an operational problem, while the removal of dissolved organic components such as volatile organic compounds is negligible. In the present work, the addition of a partially non-submerged biological granular activated carbon filter to a microfiltration lab-scale reactor was investigated. It was observed that the reactor could be operated under stable flux conditions although regular hydraulic backwashing was necessary. Preferential attached growth of nitrifiers on the activated carbon particles allowed for a complete and very stable nitrification, with permeate total ammonium nitrogen and nitrite levels below 0.2 mg/L regardless of influent concentrations. Chemical oxygen demand of the permeate averaged 5.26 mg O_2/L. Using an electronic nose, elimination of volatile compounds was assessed. The combined process resulted in complete odour removal, with permeate odour levels equalling the reference samples (demineralised water), even during periods of increased reactor load (shock load experiment). A 4.2 $\log_{10}$CFU and 3.7 $\log_{10}$CFU removal were observed for total coliforms and *E. coli*, respectively.

Keywords: membrane filtration, activated carbon, reuse, WWTP effluent, electronic nose

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[1] Redrafted after: Van Hege K., Dewettinck T., Claeys T., De Smedt G. and Verstraete W. (2002) Reclamation of treated domestic wastewater using biological membrane assisted carbon filtration. *Accepted for publication in Environmental Technology*.

INTRODUCTION

The increased incidence of water shortage during the last decades has emphasised the necessity to develop alternative ways of water conservation and explore innovative wastewater reclamation technologies (Thoeye *et al.*, 2000). An approach that has received considerable attention in recent years, is the reuse of effluent from wastewater treatment plants (WWTPs) in process water and groundwater recharge applications. In this context, it has been demonstrated that the filtrative treatment of WWTP effluent by microfiltration (MF) or ultrafiltration (UF) is highly suited for effluent polishing with the main advantages being a complete retention of colloidal and macromolecular material, compactness of the installation and low energy consumption (Ahn and Song, 1999; van Hoof *et al.*, 1998; Van Houtte *et al.*, 1998). While these processes efficiently remove particulate matter, turbidity and bacterial cells, the dissolved organic matter present in WWTP effluent is largely unaffected since retention is based on size exclusion.

Major organic constituents in WWTP effluent typically include protein, carbohydrate, humic substances, tannin, lignin as well as small amounts of surfactant (Seo and Ohgaki, 2001). Volatile organic components, originating from industrial, commercial, public and residential discharges to municipal wastewater collection systems are another important group of pollutants in WWTP effluent (Paxeus, 1996). Remediation techniques for these constituents include adsorption on granular or powdered activated carbon (GAC or PAC, respectively), air stripping and advanced oxidation processes. Satisfactory removal of organic material by MF or UF thus requires the incorporation of one of the above-mentioned unit processes.

The hybrid process resulting from the addition of activated carbon (AC) to membrane filtration reactors is a well-recognised technology and full-scale applications in drinking water production have been reported (Campos *et al.*, 2000c; Lebeau *et al.*, 1998). The advantages of such a process are fourfold: first, the AC induced sorptive organics elimination favours a higher permeate quality. For example, Campos *et al.* (2000a,b) modelled and validated the adsorptive removal of the micropollutant 4-nitrophenol in combined PAC-UF processes as a function of various operational parameters such as filtration time, membrane reactor volume, and dosing procedure. Similar work was carried out by Matsui *et al.* (2001a,b) for the herbicide simazine. In addition, it has been shown that incorporating biological activated carbon (BAC) with MF or UF further increases the refractory organics removal due to combined biodegradation and adsorption phenomena (Seo and Ohgaki, 2001; Watanabe *et al.*, 2000). Second, since AC facilitates attached growth, a more stable nitrification is observed in this type of membrane bioreactor (Kim *et al.*, 1998; Watanabe *et al.*, 2000). Besides TAN, also Mn^{2+} can be biologically oxidised by the micro-organisms inhabiting the AC particles (Suzuki *et al.*, 1998). Third, it has been demonstrated that AC pre-treatment has a beneficial effect on membrane fouling. The latter has been attributed to the formation of a more permeable cake layer on the membrane surface (Kim *et al.*, 1998) and enhanced natural organic matter removal (Tsujimoto *et al.*, 1998). A fourth advantage of combined AC - membrane filters is their ability to remove viruses by adsorption onto the activated carbon particles and subsequent rejection of these particles by membrane filtration (Seo *et al.*, 1996). Despite these apparent advantages, frequent regeneration and/or replacement of exhausted AC remains an operational problem. In PAC slurry reactors, periodic or continuous addition of substantial amounts of AC and subsequent purging is required to maintain a satisfactory permeate quality, while in GAC biofilters, filter backwashing or replacement may be necessary (Campos *et al.*, 2000b; Lebeau *et al.*, 1998; Matsui *et al.*, 2001a).

In the current work, the coupling of an *in situ* regenerative BAC filter and MF for the reclamation of treated domestic wastewater in a reactor termed Biological Membrane Assisted Carbon filtration (BioMAC), was evaluated. The removal of organic components, TAN, start-up and saturation phenomena of the GAC filter as well as the fouling tendency of the MF membranes was investigated during continuous 100-day laboratory-scale experiments. Furthermore, the elimination of organics, odorous compounds and biological constituents were assessed during a subsequent shock load experiment.

MATERIALS AND METHODS

Experimental set-up

The combined BAC-MF system consists of two unit processes, biological granular activated carbon filtration and microfiltration (Figure 1). The characteristics of the GAC and membrane material used in the process modules are summarised in Table 1.

The feed solution, stored in a 100 L feed tank, is fed into the GAC filtration column (containing 0.5 kg GAC), which is kept 30 % non-submerged by a level control directing the feed pump. After GAC filtration, the percolate is pumped to the aerated (2 L/min) membrane module. A part of the percolate is recycled over the GAC reactor (recirculation flow Q_R set at 12 L/h) and another part is microfiltered (flow Q initially set at 2 L/h), resulting in a recycle ratio Q_R/Q of 6. Transmembrane pressure (TMP) is measured by a pressure gauge and is set at a maximum of 10 kPa. The total reactor volume is 4 L, yielding an initial hydraulic retention time of 2 h.

Table 1 Characteristics of the materials used in the experimental set-up

Unit process	Parameter	Value
BAC reactor	Type	Lurgi Hydrafin CC 8 x 30
	Density (kg/m³)	480 ± 50
	Particle size (mm)	0.5 – 2
	Iodine number (mg/g)	950
Membrane module	Type	Zenon Zeeweed®
	Material	proprietary
	Surface (m²)	0.1
	Pore size (µm)	0.4

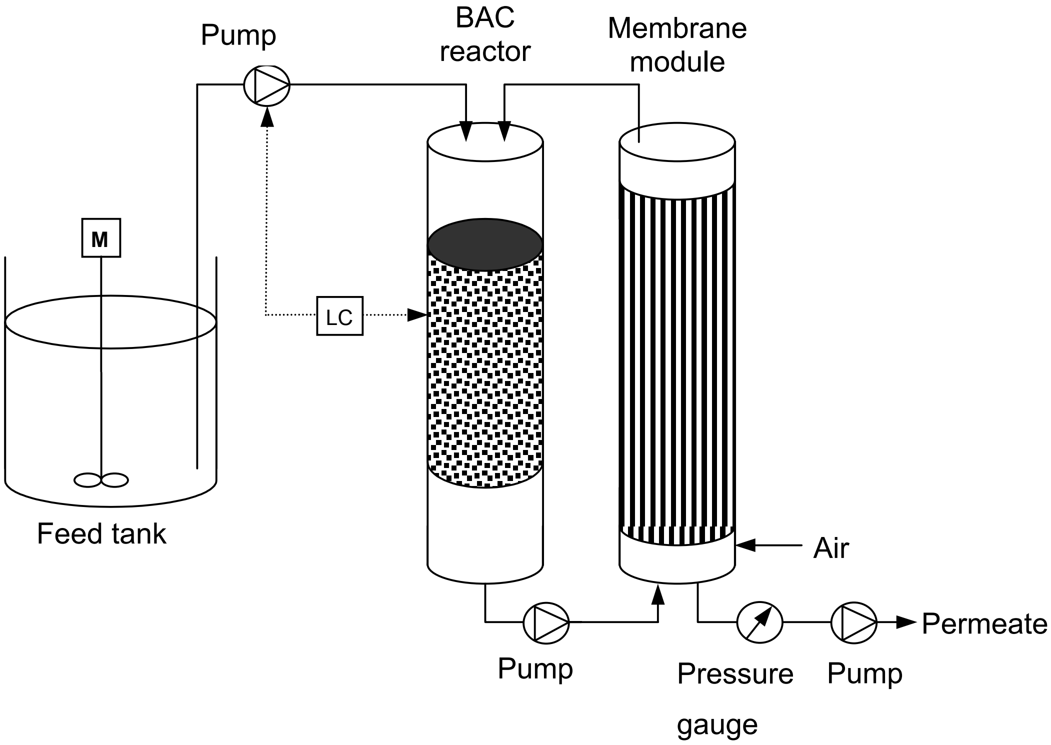

Figure 1 Scheme of the experimental set-up (M = motor, LC = Level Control)

Prior to experimental initiation, the GAC (dried at 105 °C for 24 h) is seeded with micro-organisms by circulating WWTP effluent over the filter for 3 days. The GAC filter was entirely replaced after 2400 (day 67) and again after 1400 bed volumes (day 115). The reactor was operated at room temperature (20 ± 2 °C).

WWTP effluent and influent

WWTP effluent and influent were obtained from the municipal WWTPs of Ghent and Wetteren (Belgium). Sewage treatment in both plants consists of primary sedimentation, followed by conventional activated sludge treatment and gravitational clarification. Influent and effluent were stored at 4 °C and processed within 5 days. From day 1 to 114, only WWTP effluent was fed to the reactor. On day 115, 10 L of WWTP influent was added to the feed tank (10 vol % spike) in order to obtain higher input concentrations. Subsequently, again only WWTP effluent was added to the feed tank. To avoid excessive ammonia volatilisation, pH of the reactor feed was limited to 8.0 (day 1-67) or 7.5 (day 68-129) by dosing 0.1 M H_2SO_4 (VEL, Leuven, Belgium) in the feed tank.

Physico-chemical parameters

Reactor influent as well as permeate were analysed for a number of physico-chemical parameters. Total Ammonium Nitrogen (TAN), suspended solids (SS), chemical oxygen demand (COD) and pH were determined according to the Standard Methods for the examination of water and wastewater (AHPA, 1992).

Influent and effluent nitrate, nitrite, phosphate and sulphate concentrations were determined using a Dionex DX 600 Ion Chromatograph (Dionex, Sunnyvale, USA) equipped with a conductivity detector. Operational parameters were as follows: Column AS9-HC; eluens 9mM Na_2CO_3; flow 1mL/min; sample loop 200 µL. Dissolved oxygen was measured in the recycle to the membrane module using a DO-electrode (Endress & Hauser Oxygen Probe COS 381, Brussels, Belgium).

Microbiological constituents

Plate counts were performed using the spread plate technique (reactor feed) or membrane filtration technique (permeate) (AHPA, 1992). The following agars, incubation times and temperatures were used (Atlas, 1993): Mc Conkey agar (Difco, USA), 1 day at 43 °C for *E. coli* and Violet Red Bile Agar (Oxoid, England), 1 day at 37 °C for total coliforms.

TAN volatilisation

Possible stripping of ammonia was monitored by bubbling the gaseous reactor emissions through a washing bottle containing a 1 M H_2SO_4 solution (VEL, Leuven, Belgium). In the washing bottle, gaseous ammonia is immediately hydrolysed to aqueous ammonium and the increase of the TAN concentration was measured as mentioned above. The TAN volatilisation then expressed as a percentage of the amount of TAN fed to the reactor during the time of the analysis:

$$\%\ \mathrm{TAN}_{\mathrm{volatilised}} = \frac{\left(C_{g,2} - C_{g,1}\right)V}{C_{aq}\ Q\ t} * 100 \qquad (1)$$

With ($C_{g,2}$-$C_{g,1}$) the increase in TAN concentration in the washing bottle (mg/L), V the volume of the washing bottle solution (L), C_{aq} the reactor feed TAN concentration (mg/L), Q the flow of the reactor feed (L/h) and t the time during which the reactor off-gases were sent through the washing bottle (h).

Electronic nose analyses

On the reactor influent and effluent, electronic nose measurements as described by Dewettinck *et al.* (2001b) and Van Hege *et al.* (2001) were carried out using an electronic nose (FOX 3000, Alpha M.O.S., Toulouse, France) containing 12 metal oxide sensors. Experimental details are mentioned in Chapter 4 of this dissertation.

Statistical analysis

Statistical analysis was performed using a paired 2-tailed T-test where a p value ≤ 0.01 or 0.05 was considered significant; results are shown as average ± standard deviation (SD).

RESULTS

Flux

The permeate flow was initially set at 2 L/h (corresponding to a flux of 20 L/m^2h) by regulating the permeate pump speed. Once the maximum allowed TMP of 10 kPa was reached, a flux decrease was observed as visualised in Figure 2. Backwashing with permeate (15 minutes at 4 L/h) or air (15 min at 400 kPa) on day 3, 10, 17, 21, 23, 31, 38, 42, 52 and 60 allowed for complete flux recovery during the first 60 days of operation, although a pronounced flux decrease between backwash cycles was noticed. Replacement of the GAC filter on day 67 (after 2400 bed volumes) and concomitant manual and chemical membrane cleaning (1 M HCl acid / 1 M NaOH caustic backwash at 4 L/h) led to stable operation from day 67 to day 80. Feed pump failure on day 81 gave rise to membrane exposure to ambient air; subsequent backwashing did not result in full flux recovery, indicating irreversible fouling and/or damage to the membrane module. No more backwashing was performed from day 100 on; the flux gradually decreased to 10 L/m^{2}h on day 115. The second GAC filter was replaced on day 115 after 1400 bed volumes. Subsequently, the shock load experiment was initiated.

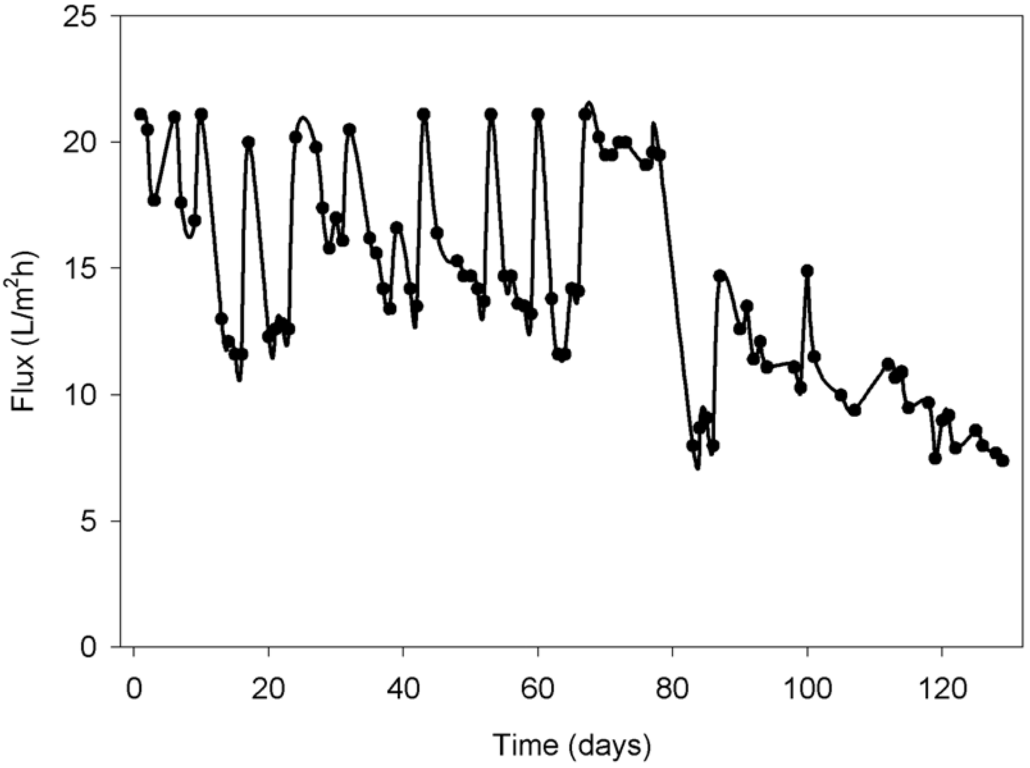

Figure 2 Evolution of the flux in the experimental set-up. On day 67 and 115, the GAC filter was replaced

Continuous run

The BioMAC reactor was operated continuously for 114 days and a number of physico-chemical parameters were measured. Results for the analyses of pH, phosphate, sulphate, TAN, nitrite, nitrate and COD are listed in Table 2.

Only for pH, TAN, nitrite, COD and sulphate a significant difference ($p \leq 0.01$) between feed and permeate could be observed. The latter was attributed to the dosing of sulphuric acid for controlling the pH of the reactor feed. No significant difference could be noticed between the input and output levels of phosphate and nitrate. In the reactor permeate, no suspended solids could be detected.

Table 2 pH, phosphate, sulphate, TAN, nitrite, nitrate and COD of the feed and permeate of the BioMAC reactor (day 1 - 114). Values are noted as average ± SD. n = number of samples. * = Significantly different from the feed, $p \leq 0.01$

Parameter	Unit	Feed	Permeate	n
pH	/	7.71 ± 0.26	8.05 ± 0.22 *	72
PO_4^{3-} -P	mg/L	0.96 ± 0.31	0.92 ± 0.30	22
SO_4^{2-} -S	mg/L	34.4 ± 4.6	47.2 ± 11.2 *	22
TAN	mg/L	0.30 ± 0.62	0.030 ± 0.026 *	47
NO_2^- -N	mg/L	0.37 ± 0.79	0.029 ± 0.050 *	36
NO_3^- -N	mg/L	3.85 ± 1.57	3.71 ± 1.46	40
COD	mg O_2/L	13.7 ± 5.0	5.26 ± 3.18 *	48
SS	mg/L	6.4 ± 1.5	Not detected (> 5)	13

Figure 3.a shows the feed and permeate concentrations for TAN. It is apparent that permeate TAN concentrations do not exceed 0.1 mg/L TAN, while from Table 2 an average TAN-removal of 90 % and a ratio $SD_{feed}/SD_{permeate}$ of 24 could be calculated. In Figure 3.b, the nitrite concentrations of the BioMAC feed and permeate are indicated. Permeate nitrite concentrations are below 0.2 mg/L and the average removal and ratio of input to output SD were found to be 92 % and 16, respectively (Table 2). While a paired T-test revealed a significant decrease in permeated TAN and nitrite content, this was not the case for the nitrate concentration. Indeed, most of the time, no notable difference could be observed between the input and output nitrate concentrations, as displayed in Figure 3.c. However, during periods of peak TAN and/or nitrite loading (for example day 85), a considerable increase in the permeate nitrate concentration was noticed.

Analysis of the gaseous phase of the reactor outlet on day 29 and 84 revealed that stripping of ammonia could be ruled out as a possible removal mechanism, as volatilisation was observed to be less than 0.5 % of total aqueous TAN input.

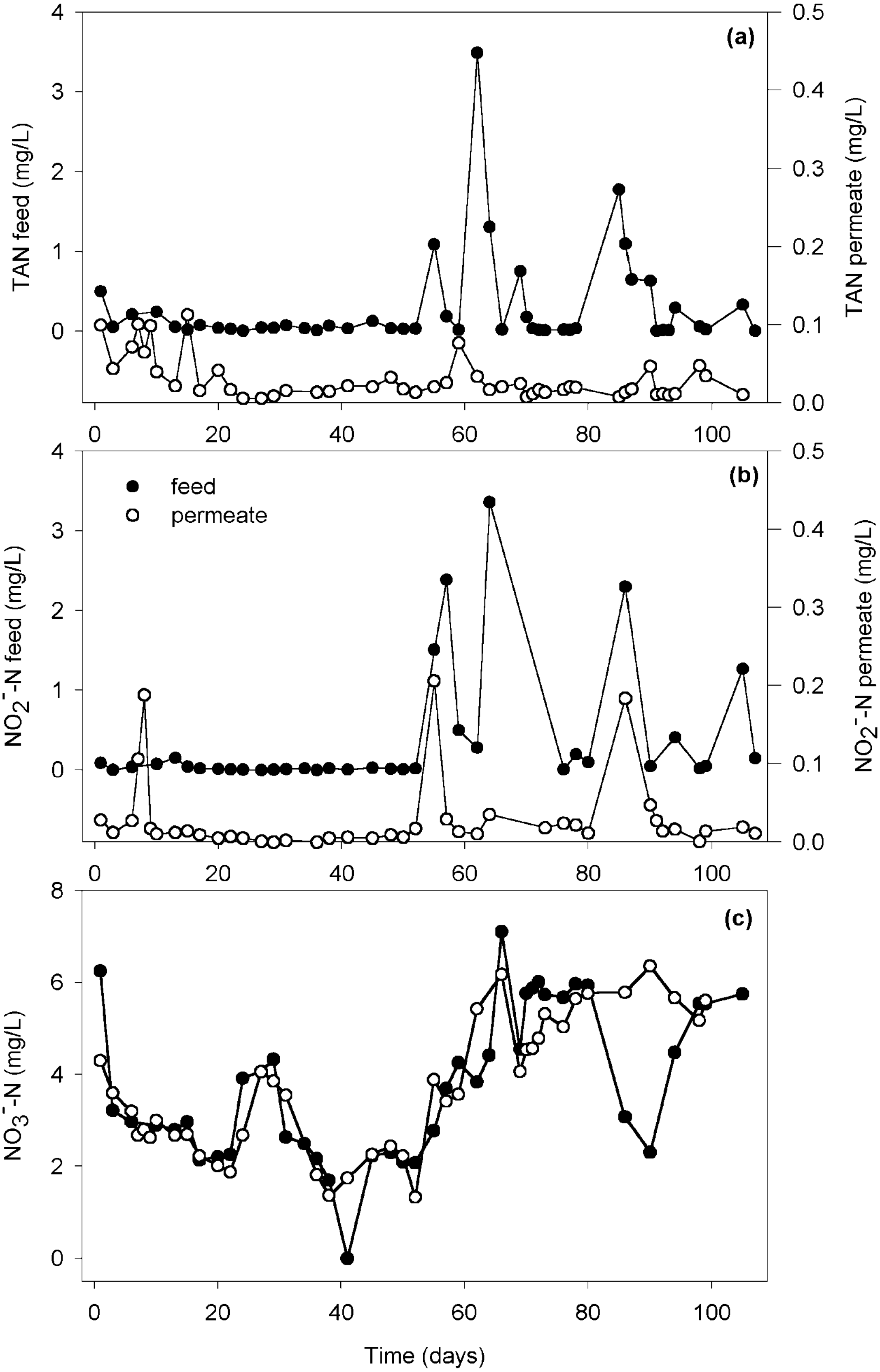

Figure 3 Evolution of (a) TAN, (b) NO_2^--N and (c) NO_3^--N concentration (mg/L) of feed and permeate of the BioMAC reactor during the continuous run (day 1 - 114)

In Figure 4, the COD concentrations of feed and permeate are depicted.

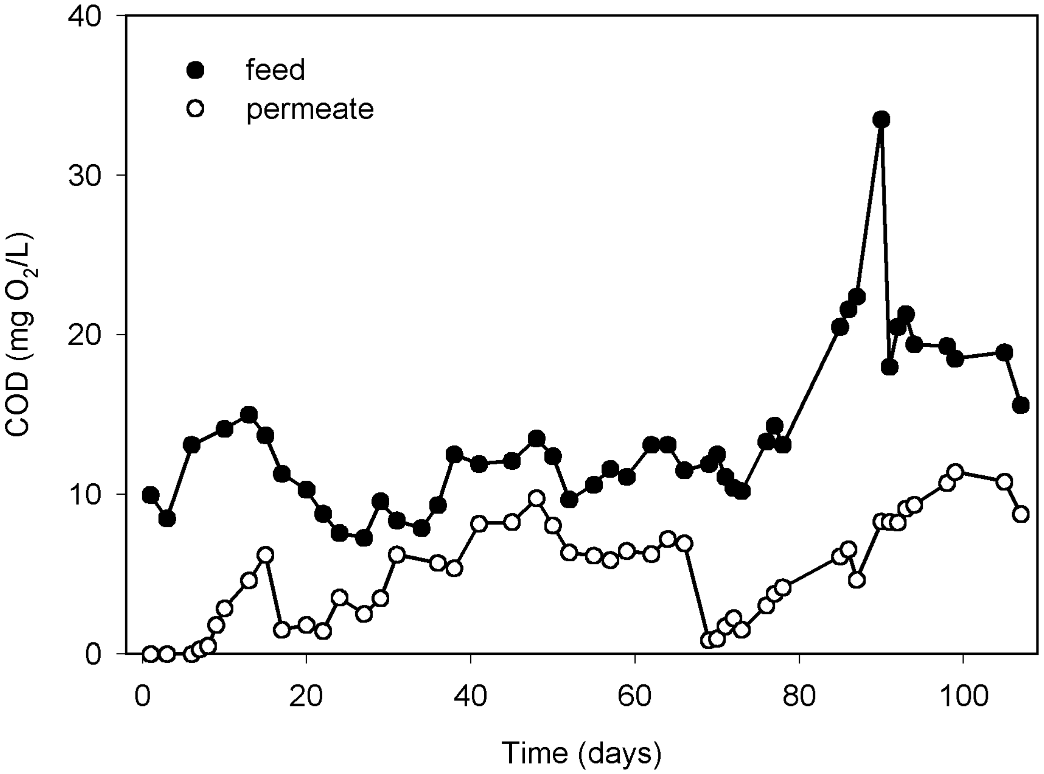

Figure 4 Evolution of COD concentration (mg O_2/L) of feed and permeate of the BioMAC reactor during the continuous run (day 1 - 114)

Initially, COD removal was complete, but from day 10 on, a gradual increase of the permeate concentration was observed (maximum permeate COD of 9.76 mg O_2/L on day 48). Replacement of the GAC filter on day 67 brought about a sharp decrease in permeate COD concentration, but a subsequent increase was noticed (maximal permeate COD of 11.4 mg O_2/L on day 99). An average COD-removal of 39 % was calculated and the ratio $SD_{feed}/SD_{permeate}$ was found to be 1.6. DO levels after the biological GAC filtration were around 5.3 mg/L.

Shock Load Experiment

On day 115, the GAC filter was entirely replaced and the reactor was fed with spiked WWTP effluent (WWTP influent spike). From day 115 to 129, the elimination of organics, odorous compounds and biological constituents was measured to evaluate the reactor performance during abrupt variations of input concentrations. The results are indicated in Table 3 and Figure 5.

Table 3 Measured values of the SOP_{av} (feed, permeate and reference), COD, total coliforms and *E. coli* (feed and permeate) of the BioMAC reactor during the shock load experiment (day 115 to 129). Values are noted as average ± SD. n = number of samples. * = Significantly different from the feed, $p \leq 0.01$. ** = Significantly different from the feed, $p \leq 0.05$

Parameter	Unit	Value	n
SOP $_{av, feed}$	/L	329 ± 48	10
SOP $_{av, permeate}$	/L	250 ± 61 *	10
SOP $_{av, reference}$	/L	233 ± 38	10
COD $_{feed}$	mg O_2/L	36.3 ± 21.1	9
COD $_{permeate}$	mg O_2/L	4.86 ± 1.00 *	9
Total coliforms $_{feed}$	log_{10}CFU/100 mL	5.4 ± 0.5	3
Total coliforms $_{permeate}$	log_{10}CFU/100 mL	1.24 ± 0.83 **	3
E. coli $_{feed}$	log_{10}CFU/100 mL	4.3 ± 1.3	3
E. coli $_{permeate}$	log_{10}CFU/100 mL	0.55 ± 0.95 **	3

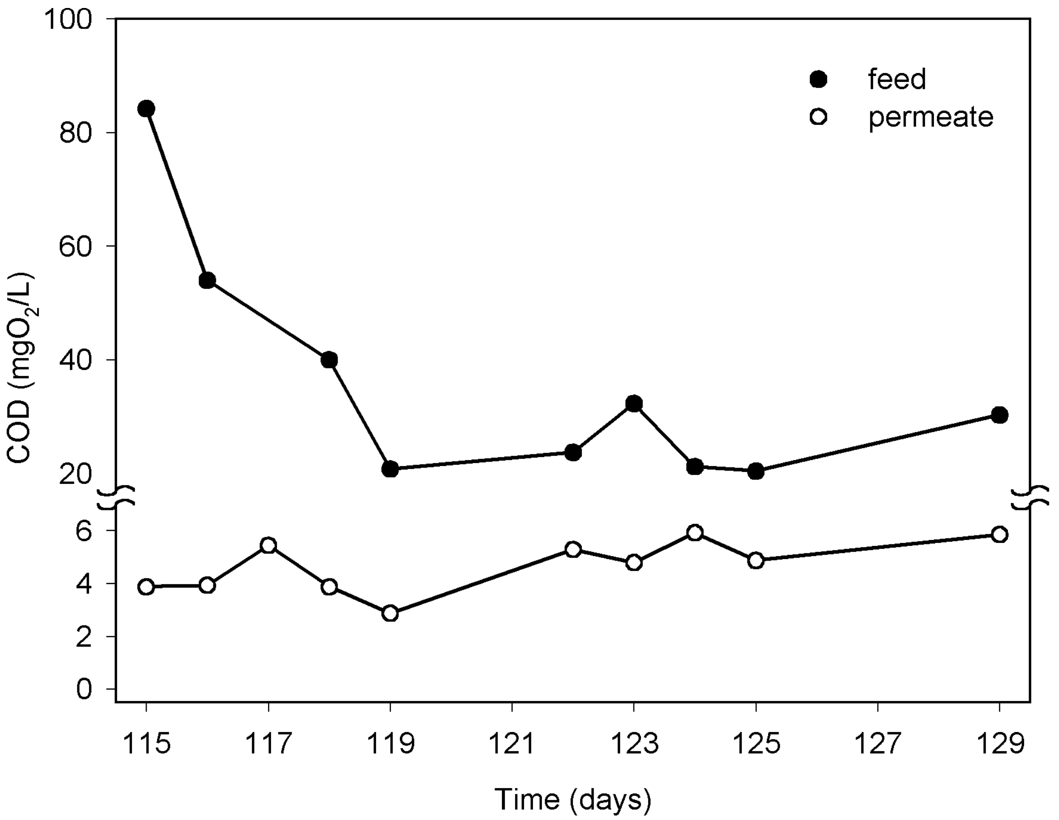

Figure 5 Evolution of COD concentration (mg O_2/L) of feed and permeate of the BioMAC reactor during the shock load experiment (day 115 to 129)

Figure 5 reveals that addition of WWTP influent to the reactor feed gave rise to a major COD increase. However, subsequent dilution by the addition of WWTP effluent to the feed tank resulted in a COD level of around 20 mg O_2/L (day 119). The average permeate COD was significantly lower ($p \leq 0.01$), averaging 4.68 mg O_2/L. The ratio $SD_{feed}/SD_{permeate}$ was 21 and an average COD removal of 85 % was noticed.

During the shock load experiment, the reactor feed and permeate were assessed with regard to total coliform and *E.coli* removal efficiency. The average feed and permeate concentrations are expressed as $\log_{10}$CFU per 100 mL of sample and are shown in Table 3; a significant decrease is noted ($p \leq 0.05$). An average removal of 4.2 and 3.7 $\log_{10}$CFU was computed for total coliforms and *E. coli*, respectively. Furthermore, in most permeate samples, no bacterial indicator organisms could be detected in 100 mL of sample (membrane filtration technique). More importantly, total coliform permeate levels were observed to be below a level of 200 CFU /100 mL

The fate of odorous compounds was also monitored and expressed as SOP_{av}-values (Table 3). The SOP_{av}-values, which are an indication of the odour quantity, of both the permeate and reference samples are significantly lower ($p \leq 0.01$) than the feed SOP_{av}. On the other hand, the difference between the odour quantity of the permeate and reference (demineralised water) was observed to be statistically insignificant ($p \leq 0.01$). In Figures 6.a and b, the odour quality of the feed, permeate and reference is visualised in radarplots for day 115 and 125, respectively. Although the sample of day 125 displays a lower odour quantity, in both cases a considerable difference between the odour pattern of the feed samples and that of the reference is noticed. On the other hand, both permeate samples bear a fairly high resemblance to the reference fingerprints.

DISCUSSION

In the present study, the possibility to reclaim WWTP effluent by a biological membrane assisted carbon filtration (BioMAC) reactor has been explored. The positive influence of GAC or PAC pre-treatment on subsequent membrane filtration kinetics has been extensively documented (Kim *et al.*, 1998; Seo and Ohgaki, 2001; Tsujimoto *et al.*, 1998; van der Hoek *et al.*, 1999). Tsujimoto *et al.* (1998) found that biological GAC pre-filtration was effective in preventing irreversible fouling, thus considerably decreasing the chemical cleaning frequency.

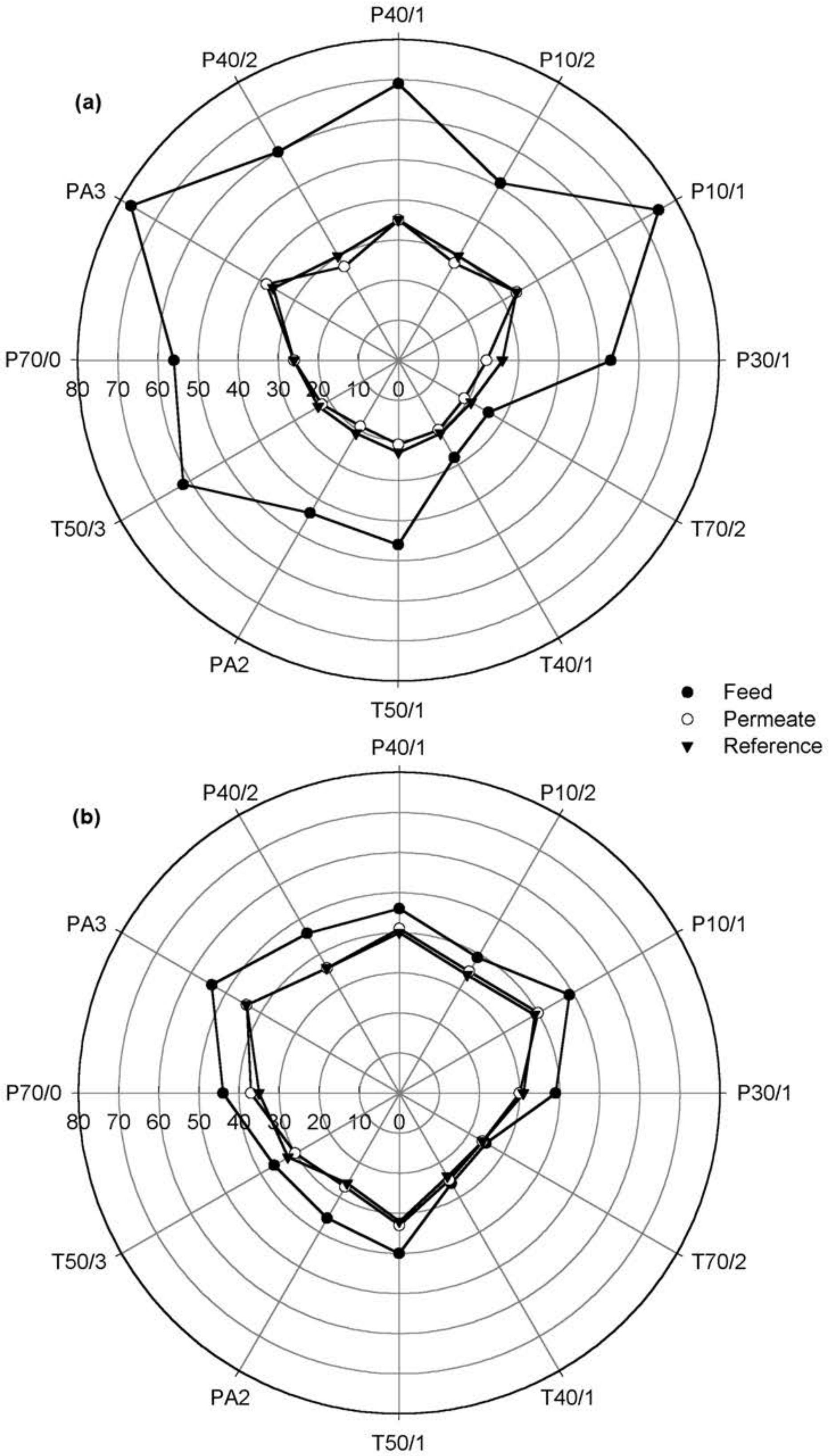

Figure 6 Radar plot of the odour profile of the feed, permeate of the BioMAC reactor on day 115 (a) and day 125 (b). The 12 axes correspond to the 12 different sensors of the FOX 3000 electronic nose. The data points are the relative change in electrical resistance (SOP) of each sensor

From Figure 2, it can be seen that initial backwash operations resulted in full flux recovery, supporting the hypothesis of Tsujimoto *et al.* (1998). Moreover, after GAC filter replacement on day 67, membrane operation was stable up to day 80, indicating a severe reduction in fouling tendency by elevated sorptive organic matter removal as described by van der Hoek *et al.* (1999). Membrane exposure on day 81 and subsequent absence of backwashing operations (be it hydraulic or chemical) resulted in flux decrease to around 8 L/m^{2}h on day 115, after which the flux stabilised. The existence of a flux value below which there is no substantial fouling has been reported in literature where it has been termed critical flux (Howell, 1995).

The removal of organic matter in BAC filters is known to be a synergetic combination of physical sorption and biodegradation: on the one hand, biodegradation results in recovery of the adsorptive capacity of the AC, while on the other hand physical sorption can reduce the toxicity of wastewater contaminants towards micro-organisms (Sirotkin *et al.*, 2001). It is recognised that the effect of the influent oxygen concentration on these phenomena is of paramount importance (Scholz and Martin, 1997; Warta *et al.*, 1995). In the BioMAC reactor, a part of the GAC filter is kept non-submerged, so saturated oxic conditions can be expected. In addition to this, oxygen saturated water is recycled over the filter resulting in an average DO of 5.3 mg O_2/L after the GAC filtration. Warta *et al.* (1995) demonstrated that such high concentrations of molecular oxygen induce surface catalysed polymerisation reactions of natural organic matter which result in an appreciable increase of the adsorptive capacity of carbon. According to Miserez *et al.* (1999) the above-mentioned conditions stimulate the growth of a thin biofilm of a highly performant interactive biomass on the GAC. Consequently, the enhanced physical sorption and pronounced biodegradation can substantially extend the period between successive reactivation or replacement. In the reactor under scrutiny, the average permeate COD concentrations were 5.26 and 4.86 mg O_2/L for the continuous run and the shock load experiment, respectively. Nevertheless, the COD concentration exceeds 10 mg O_2/L from day 98 to day 105. From this it is clear that, despite the inherent regenerative capacities of the oxygen saturated BAC filter, a periodic *ex situ* GAC replacement or regeneration remains inevitable.

Based on the experimental data (*i.e.* the unsubstantial nitrite or TAN concentrations in the permeate, the absence of ammonia in the gaseous phase and the increase of the permeate nitrate content during TAN input peaks), the extent of nitrification was considered to be complete. According to Kim *et al.* (1998), the complete and stable nitrification process is due to the fact that activated carbon particles favour the attached growth of nitrifying bacteria. In their study of the PAC/MF process for drinking water production from surface water, Lebeau *et al.* (1998) noted an excellent biomass resistance to abrupt variations of TAN concentration. In the present work, this exceptional stability is evidenced by the ratio of the standard deviation of influent to effluent concentration for nitrite and TAN (24 and 16 respectively), as a ratio substantially larger than 1 is considered to be indicative for stable process performance (Jefferson *et al.*, 2000).

Because of the strong public perception regarding the presence of tastes and odours in water, successful reclamation of WWTP effluent – even for purposes other than drinking water production - necessitates the elimination of these components. In recent publications, the measurement of volatile (odorous) compounds in sewage treatment works using an electronic nose has been described (Dewettinck *et al.*, 2001b; Gostelow *et al.*, 2001). During the shock load experiment, it was shown that the BioMAC process successfully removed odorous components present in the feed solution. The electronic nose radar plots (Figures 6.a and 6.b) as well as the SOP_{av} values indicate that the permeate possesses essentially the same odour quality and quantity as the reference samples. In their assessment of taste and odour removal, Laine *et al.* (2000) found that low pressure membrane processes solely could not reduce taste and odour levels, but that the combination PAC-UF was effective for taste and odour control. In this respect, it seems logical that the odour removal in the BioMAC reactor was achieved by the biological GAC filter.

Membrane processes are known to posses a certain disinfecting capacity (Madaeni, 1999). For example, Johnson *et al.* (1997) observed a 6.1 $\log_{10}$ faecal coliform removal in a MF pilot scale plant, while Gander *et al.* (2000b) demonstrated a 5 $\log_{10}$ elimination of total coliforms in a polypropylene membrane bioreactor. In addition, Madsen (1987) noted a 8 $\log_{10}$ rejection of *E. coli* by MF. The relatively small observed retention (4.2 $\log_{10}$CFU and 3.7 $\log_{10}$CFU for total coliforms and *E. coli*, respectively) can, however, be explained since it is known that membrane retentivity depends on the magnitude of the organism challenge (Madaeni, 1999). Indeed, in their pilot study of a PAC/MF reactor with in-line coagulation, Lebeau *et al.* (1998) observed a coliform retentivity ranging from 2 to 4 $\log_{10}$CFU, depending on the micro-organism content of the raw water.

When comparing the experimental data with the United States Environmental Protection Agency Guidelines for water reuse (US EPA, 1992), a reclaimed water is obtained that as such is suitable for non-contact reuse applications such as cooling water, agricultural irrigation of crops to be commercially processed and restricted access irrigation. Although the BioMAC reactor permeate is essentially free of SS and odour, unrestricted potable reuse would necessitate additional disinfection to comply with the Guideline's faecal coliform regulation (*i.e.* no faecal coliforms in 100 mL).

CONCLUSIONS

A combined biological granular activated carbon and microfiltration process was evaluated with regard to fouling tendency; removal of COD, TAN, odorous components, biological constituents and shock load resistance. For the WWTP effluents under scrutiny, it can be concluded that the BioMAC process, which includes an enhanced *in situ* regenerative BAC filter, allows for :

- Stable hydraulic operation, either above (combined with regular hydraulic backwash cycles) or below subcritical flux conditions (*i.e.* 8 L/m^2h).
- Stable and complete nitrification with residual TAN and nitrite levels below 0.2 mg/L regardless of influent peak levels.
- Removal of COD to levels lower than 10 mg O_2/L, provided the AC filter is regularly regenerated.
- Odour removal to a level comparable to the reference samples.
- At least a 3 $\log_{10}$CFU rejection of total coliforms and *E. coli*.

According to the US EPA guidelines for water reuse, the produced water would be suited for unrestricted nonpotable reuse after additional disinfection.

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CHAPTER 6

END-OF-PIPE MEMBRANE CONCENTRATE TREATMENT[1]

ABSTRACT

The cost of disposal or treatment of membrane concentrates is rapidly becoming one of the main factors influencing the feasibility of any membrane application. In the present work, the results of biological and electrochemical treatment of reverse osmosis membrane concentrates are discussed. As biological tests using the BioMAC reactor illustrated the very poor biodegradability of the RO brine, research efforts were focussed on electrolysis as a novel treatment approach. Four different materials have been tested as anode for a number of brine samples in a one-compartment electrolytic cell in galvanostatic mode. It was found that PbO_2 and SnO_2 anodes initiated electrochemical precipitation through an increase of the pH. Boron-doped diamond and RuO_2 coated Ti anodes successfully oxidised the pollutants in the brine and a linear removal of TAN and COD was observed during the first phase of oxidation. Oxidation was predominantly achieved through indirect hypochlorite bulk oxidation; the higher oxidation rate and extent for the boron-doped diamond anode was attributed to the higher selectivity and activity of the latter. Overall performance of the boron-doped diamond electrode was higher than for RuO_2: higher rates for TAN (17.9 vs. 13.5 mg/Ah) and COD (74.5 vs. 20.0 mg/Ah) removal as well as higher overall current efficiencies (35.2 vs. 14.5 %) were noted. Extensive colour removal was observed for both anodes (> 90 % decrease in absorbency at 455 nm).

Keywords: RO retentate, reject brine, electrolysis, electrochemical treatment, COD removal, TAN removal

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[1] Redrafted after

Van Hege K., Verhaege M. and Verstraete W. (2002) Indirect electrochemical oxidation of reverse osmosis membrane concentrates at boron-doped diamond electrodes. *Electrochemistry Communications*, **4** (4), 296-300.

Van Hege K., Verhaege M. and Verstraete W. (2002) Electro-oxidative abatement of reverse osmosis membrane concentrates. *Submitted.*

INTRODUCTION

In the last decades, numerous full-scale drinking water production centres and reuse facilities employing membrane technology have been planned and commissioned to comply with the ever-increasing world-wide demand for pristine, unpolluted water. The filtrative treatment of waters ranging from wastewater treatment plant effluent to seawater has proven to be a reliable and sustainable reclamation technology (Weber and LeBoeuf, 1999). While these processes have many attractive features, the cost of disposal or treatment of the filter retentate (also referred to as membrane concentrate or reject brine) is rapidly becoming one of the main factors influencing the feasibility of any membrane application (Squire *et al.*, 1996).

According to Ahmed *et al.* (2001), the cost of concentrate discharge can range from 5 to 33 % of the total treatment cost. Disposal options, such as direct surface water discharge or deep well injection, are being restricted because of increased environmental awareness and more stringent regulations. Discharge into the sewer system may be a viable option, but the brine composition could adversely affect the sewage treatment processes while the augmentation of the total dissolved solids content of the final effluent may render it inept for irrigation purposes (Ahmed *et al.*, 2001; Squire, 2000). The use of lined evaporation ponds is restricted to countries with hot and dry climate conditions coupled to the availability of land at low cost (Ahmed *et al.*, 2000).

The chemical characteristics of membrane concentrates such as reverse osmosis (RO) retentate have a profound impact on the possible treatment options. Biological treatment may be impeded by the inhibitory effect of the high salinity on the microbial growth or prove to be inefficient because of the recalcitrance or biotoxicity of the organic constituents. Several other treatment approaches have been reported: for example, Balanosky *et al.* (1999) used an immobilised Fenton reagent to degrade the membrane concentrate originating from the nanofiltration of a textile effluent. Borsani *et al.* (1996) applied a three-stage flash evaporation and final crystallisation, while Facey and Smith (2001) introduced freeze thawing to further reduce the volume of contaminated waste concentrates. However, all these processes are inefficient for the removal of TAN in the retentate.

In the present study, advanced biological treatment using the BioMAC reactor is proposed as a possible abatement technology for RO concentrates (Van Hege *et al.*, 2002). The efficiency of the reactor with regard to the removal of COD and nitrogen compounds was assessed in a 60-day continuous run.

Second, electrochemistry is introduced as a novel treatment strategy for RO membrane concentrate. Electro-oxidation has been successfully implemented for the abatement of hard-to-treat wastes such as landfill leachate (Chiang *et al.*, 1995; Wang *et al.*, 2001), textile effluent (Vlyssides *et al.*, 2000) and wastewater containing polyaromatic organic pollutants (Panizza *et al.*, 2000a). Particularly with regard to RO brine, it has several advantageous features. First, the elevated salinity of RO concentrate ensures an excellent electric conductivity that could lower the energy consumption (Lin *et al.*, 1998). Second, the high chloride content could facilitate indirect bulk oxidation through the electrogeneration of strong oxidants such as hypochlorite (Chiang *et al.*, 1995). Third, oxidation of TAN and recalcitrant organics can be accomplished simultaneously (Wang *et al.*, 2001). The efficiency of electro-oxidative abatement was assessed in a number of batch experiments.

MATERIALS AND METHODS

BioMAC reactor

For the biological treatment, a Biological Membrane Assisted Carbon Filtration (BioMAC) reactor consisting of two unit processes, biological granular activated carbon filtration and microfiltration, was used. The experimental set-up and materials used have been described by Van Hege *et al.* (2002) and more detail is also given in Chapter 5 of this dissertation. The reactor was operated continuously for 60 days; the activated carbon filter was replaced entirely on day 21 and partially (50 %) on day 34.

Electrode material

Boron-doped diamond (BDD) electrodes, RuO_2 coated Ti sheets (Magneto-Chemie B.V., Schiedam, The Netherlands) and plain Ti sheets (Good Fellow, United Kingdom) were used as received. The BDD electrodes consisted of a thin (2-7 μm) highly conductive (< 0.1 Ωcm) boron-doped diamond film deposited on a Nb substrate using the hot filament chemical vapour deposition technique (HF CVD) from a gaseous feed of methane and a boron dopant in hydrogen. PbO_2 and SnO_2 electrodes were prepared by *in situ* electro-oxidation of Pb and Sn sheets under the following operational parameters: electrolyte 2 M H_2SO_4; current density 40 mA/cm²; electrolysis time 30 min.

Apparatus

Cyclic voltammetry was carried out in a conventional three-electrode cell using a computer controlled EG & G 263 A potentiostat. BDD, RuO_2 or Ti was used as the working electrode, Ag/AgCl as the reference electrode and Pt as the counter electrode. The exposed apparent area of the working electrode was 1 cm² and the scan rate was fixed at 5 mV/s. Voltammetry took place at room temperature in a stirred reactor containing 250 mL electrolyte.

Bulk electrolysis was performed in a one-compartment electrolytic flow cell in galvanostatic mode (Figure 1). Ti was used as the cathode; all electrodes were rectangular with a geometric area of 50 cm² each. The inter-electrode gap was 10 mm and the cell volume 200 mL. The electrolyte (1 L) was stored in a stirred vessel and circulated through the electrolytic cell by a peristaltic pump at a flow rate of 15 L/h. Electrolysis time was limited to 120 minutes and was carried out at different currents (0.5, 1 and 1.5 A). All experiments were performed at room temperature (20 ± 2 °C).

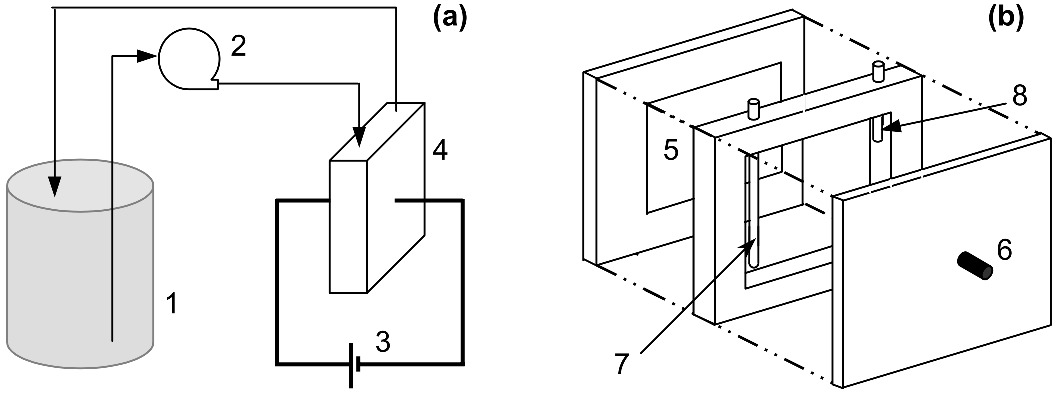

Figure 1 Scheme of the equipment used for the bulk oxidation experiments. (a) set-up used: (1) stirred vessel, (2) peristaltic pump, (3) power supply, (4) electrochemical cell. (b) Detail of the electrochemical cell: (5) anode, (6) cathode electrical connection, (7) electrolyte inlet, (8) electrolyte outlet

Scanning electron microscopy

Scanning electron microscopy was performed using a SEM (Philips XL30, The Netherlands) equipped with an EDAX® energy dispersive X-ray (EDX) spectrometer.

RO concentrate

The RO membrane concentrate samples were obtained from a 2-stage Dow Filmtech RO pilot installation further processing wastewater treatment plant effluent (Belgium). The samples were collected and stored at 4 °C in closed vessels prior to experimental initiation.

During a first period, the RO concentrate originated from effluent of a purely domestic wastewater treatment plant. This concentrate was used in the biological experiments from day 1 - 20. During a second experimental period, the pilot installation purified effluent of a mixed domestic and textile origin; this was used in the biological test from day 21 on and in the electrochemical batch experiments. More specifically, the characteristics of the 3 brine samples used for the electrolytic batch experiments are summarised in Table 1.

Table 1 Characterisation of the 3 RO brine samples used in the bulk electrolysis experiments

Parameter	Unit	Sample 1	Sample 2	Sample 3
Sampling date	/	28 8 2001	13 9 2001	3 10 2001
pH	/	8.74	7.91	8.05
EC	µS/cm	5060	5290	3990
Cl^-	mg/L	777	804	595
ClO_3^--Cl	mg/L	3.03	3.39	1.16
Ca^{2+}	mg/L	126	109	208
Mg^{2+}	mg/L	40.2	23.9	32.3
Active Chlorine	mg/L	< 0.01	< 0.01	0.07
COD	mg O_2/L	151	218	171
TAN	mg N/L	31.0	35.4	37.6
Absorbency (455 nm)	cm^{-1}	0.1463	0.2433	0.1340

Physico-chemical parameters

TAN, electrical conductivity (EC), COD and pH were determined according to the Standard Methods for the examination of water and wastewater (AHPA, 1992). Soluble magnesium, calcium and lead were quantified by flame atomic absorption spectroscopy (Perkin Elmer 3110, Überlingen, Germany) after filtration through a 0.22 µm filter and acidification. Precipitated calcium and magnesium compounds, expressed as mg/g dry matter, were analysed using the same method after drying and destruction according to the Standard Methods (AHPA, 1992). Nitrite, nitrate, chloride and chlorate concentrations were monitored using a Dionex DX 600 Ion Chromatograph (Dionex, Sunnyvale, USA) equipped with a conductivity detector. Operational parameters were as follows: Column AS9-HC; eluens 9mM Na_2CO_3; flow 1mL/min; sample loop 200 µL. Active chlorine, defined as the sum of aqueous Cl_2, HClO and OCl^-, was measured by a chlorine test (Aquaquant 1.14431.0001, Merck, Belgium). Absorbency was recorded at different wavelengths using a Uvikon 220 spectrophotometer with 1 cm optical path length. Demineralised water was used as a reference.

Statistical analysis

Statistical analysis was performed using a paired 2-tailed T-test where a p value ≤ 0.05 was considered significant; results are shown as average ± standard deviation (SD).

RESULTS

Biological treatment

During the 60-day continuous test, the pH of the feed (8.28 ± 0.08) was not significantly different form the reactor permeate (8.38 ± 0.10) (n = 24).

Figure 2 depicts the evolution of nitrate, nitrite and TAN in the feed and permeate of the BioMAC reactor. During the first experimental period (day 0 - 20), more TAN is removed as the experiment proceeds. Throughout the first 10 days of the experiment, the permeate nitrite concentrations exceed those of the feed, but from day 12 on, nitrite permeate concentrations are below the feed concentration. Nitrate concentrations are around 25 mg/L NO_3^- -N and permeate concentrations exceed those of the feed from day 8 on. After replacement of the GAC filter (at the start of the second experimental period, day 21), higher TAN levels are fed into the reactor from day 25 on. At first, the high permeate TAN levels reveal there is no removal, but from day 32 on, the TAN removal starts to pick up and it can be seen from Figure 2.a and 2.b that this initially gives rise to the formation of nitrite. This is further confirmed by Figure 2.c as no difference in feed and permeate nitrate concentration is observed. TAN is completely removed from day 40 and nitrite from day 55. Over the experimental period, the sum of the nitrite, nitrate and TAN concentrations of the reactor feed (37.5 ± 6.3 mg N/L) was not significantly different from the nitrogen content of the permeate (34.5 ± 6.1 mg N /L) (n = 22).

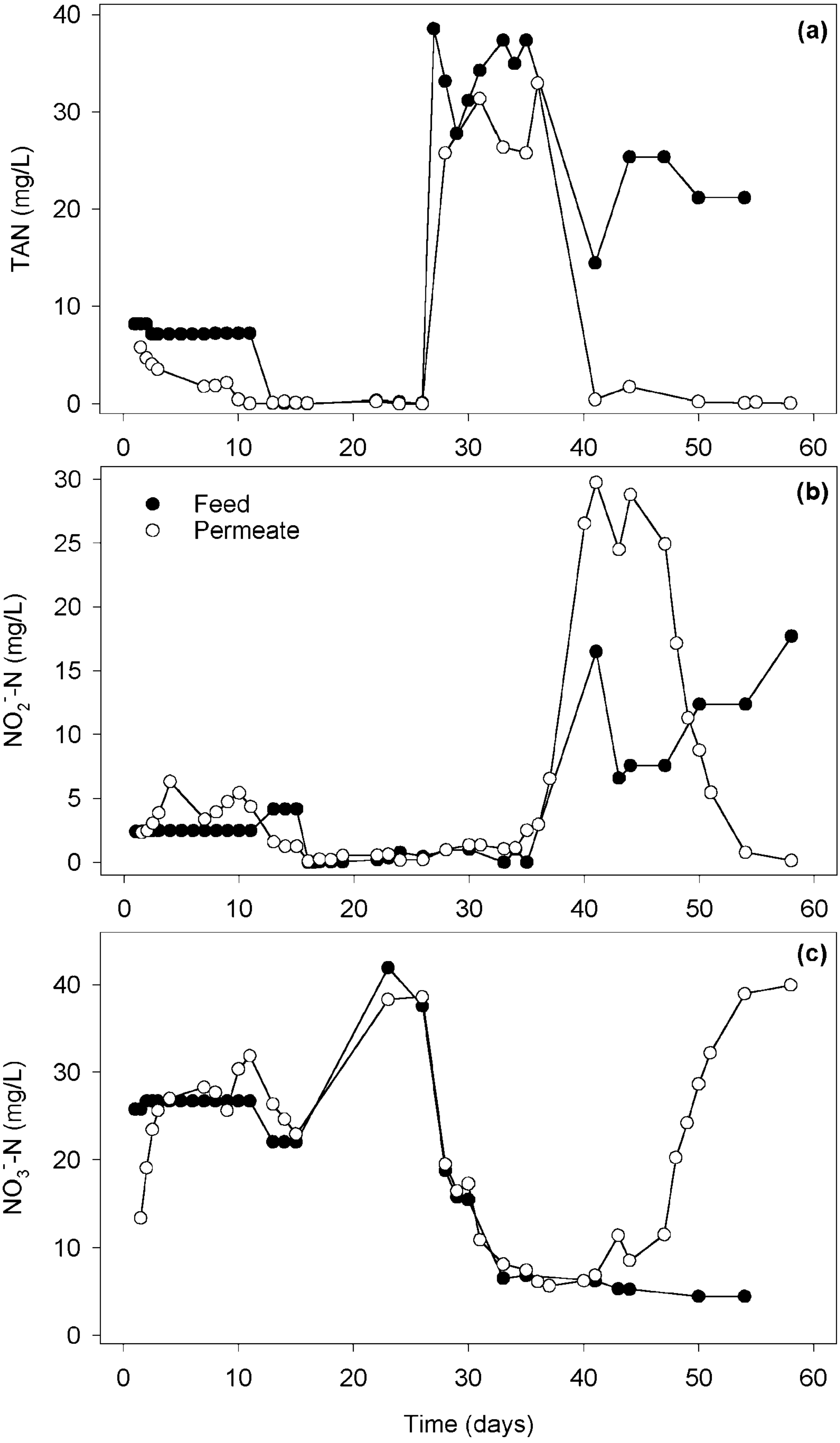

Figure 2 Evolution of (a) TAN (b) NO_2^--N and (c) NO_3^--N concentration (mg/L) of RO feed and permeate of the BioMAC reactor during the continuous run (day 1 - 60)

In Figure 3, the COD of the RO concentrate feed and the BioMAC permeate are indicated. Initially, COD removal is high (around 70 %), but after 1 week the removal decreases to 40 %. During the first 20 days of the experimental run, the average COD removal is 55 %.

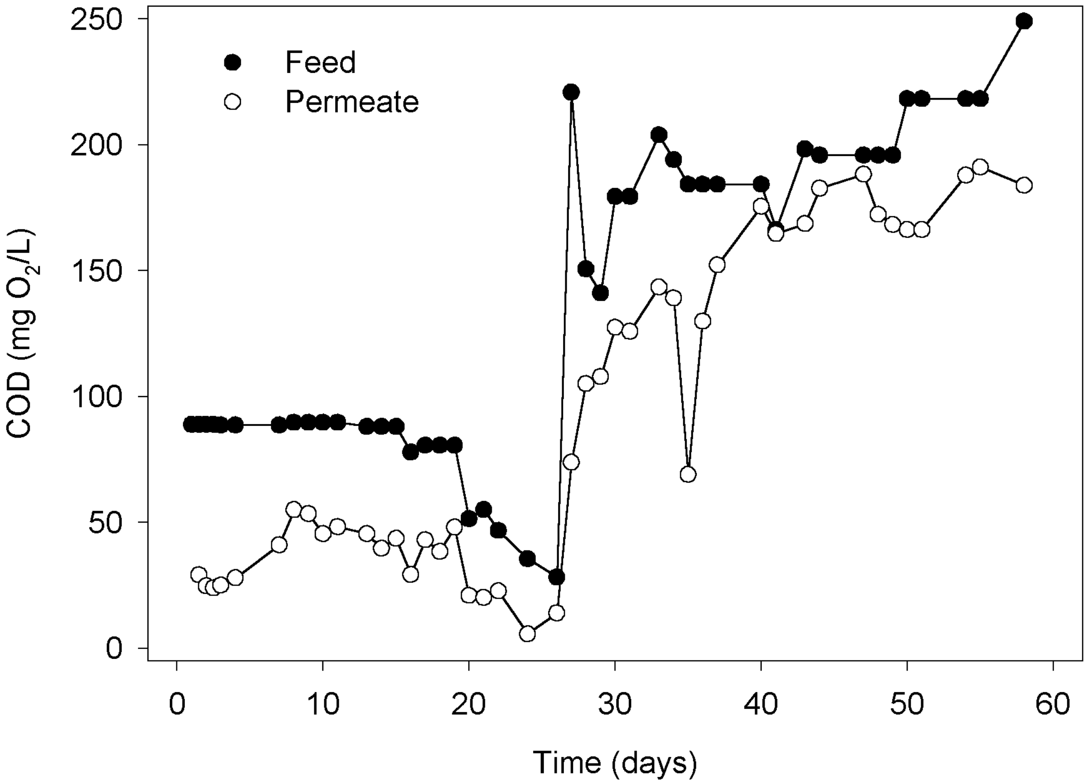

Figure 3 Evolution of COD concentration (mg O_2/L) of the RO concentrate feed and permeate of the BioMAC reactor during the continuous run (day 1 - 60)

After replacement of the GAC filter on day 21, RO concentrate originating from mixed domestic and textile effluent is used as a feed. This results in a drastic decrease of the performance of the reactor: the removal efficiency drops to 30 % after about 10 days of operation (day 30). Replacement of 50 % of the GAC filter on day 34 results in a sharp drop in the permeate COD, but this effects lasts only a couple of days. From day 40 on, the removal efficiency is very low but seems to increase towards day 60. During the second experimental period, on average only 30 % of the COD is removed.

Electrode characterisation

Figure 4 shows 2 consecutive voltammograms obtained for BDD, Ti and RuO_2 electrodes in the potential region between 0 and 4 V (vs. Standard Hydrogen Electrode, SHE) with a scan rate of 5 mV/s.

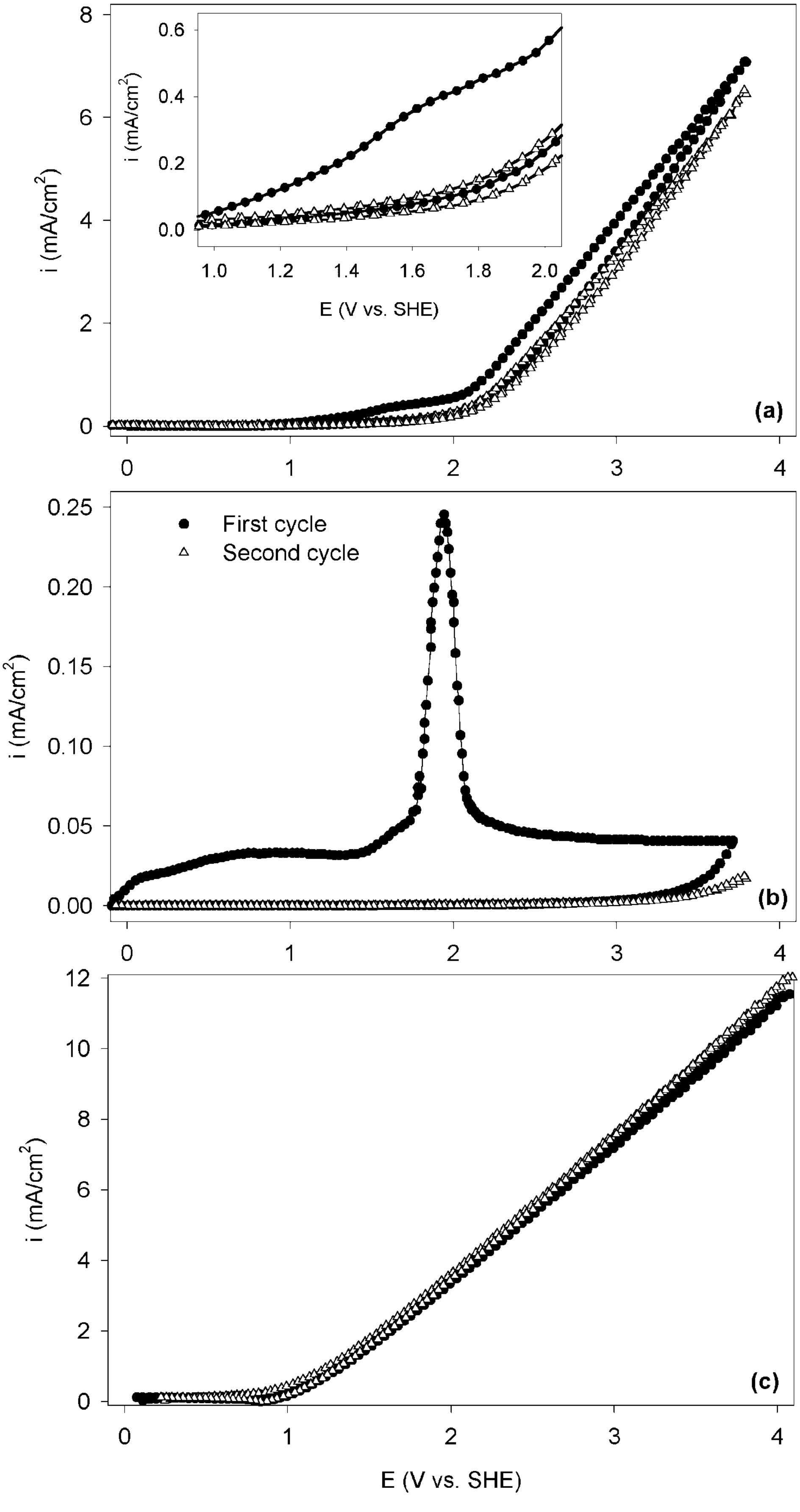

Figure 4 Cyclic voltammograms on (a) BDD, (b) Ti and (c) RuO_2 electrodes in RO brine sample 3 in the potential region between 0 and 4 V vs. SHE

The BDD anode displays a small anodic current peak at 1.6 V (Figure 4.a, inset), which disappears nearly completely in the second cycle. During the first cycle, a small peak is also observed for the Ti electrode. After one scan, the resistance of the Ti electrode considerably increases, indicating that the electrode becomes passivated (*i.e.* a TiO_2 layer is formed). In the case of the RuO_2 coated Ti anode, both cyclic voltammograms overlap, while the current is higher than for the BDD or Ti electrodes.

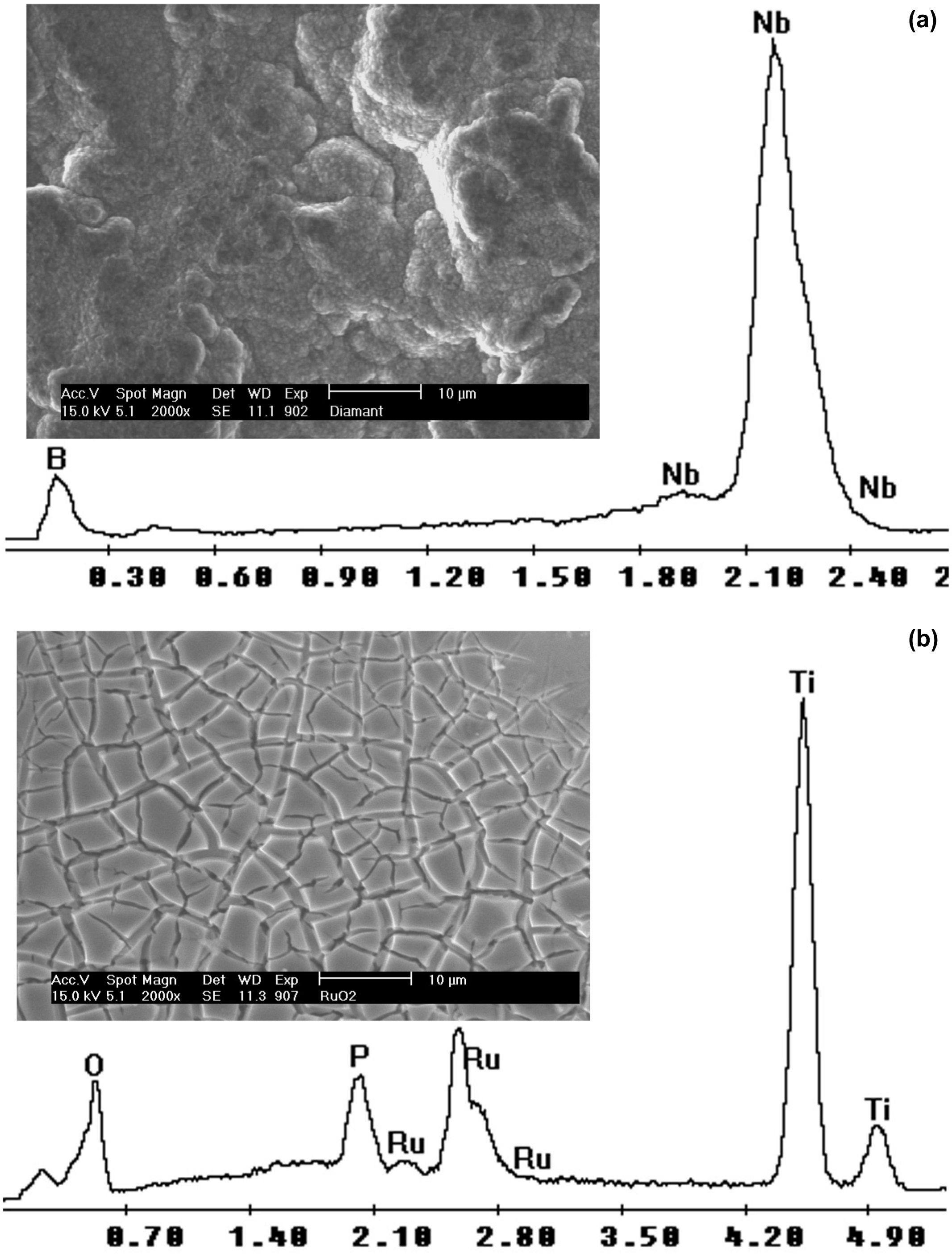

Figure 5 EDX and SEM micrographs for (a) BDD and (b) RuO_2 electrode surface.

Figures 5.a and 5.b represent the EDX spectra and SEM micrographs of the BDD and RuO_2 surface, respectively. The BDD surface has an amourphous appearance; the EDX spectrum shows a small peak origination from the doping element boron and a larger peak for the niobium substrate material (The settings of the EDX did not allow for carbon to be detected). From the EDX of the RuO_2 electrode surface, peaks for ruthenium, oxygen, phosphorus and titanium are identified.

Batch electrolytic experiments

A first set of experiments was performed to screen for a suitable anode material. Figure 6 depicts the evolution of the pH during the electrolysis of brine sample 3 using different anode materials (galvanostatic conditions, 20 mA/cm²).

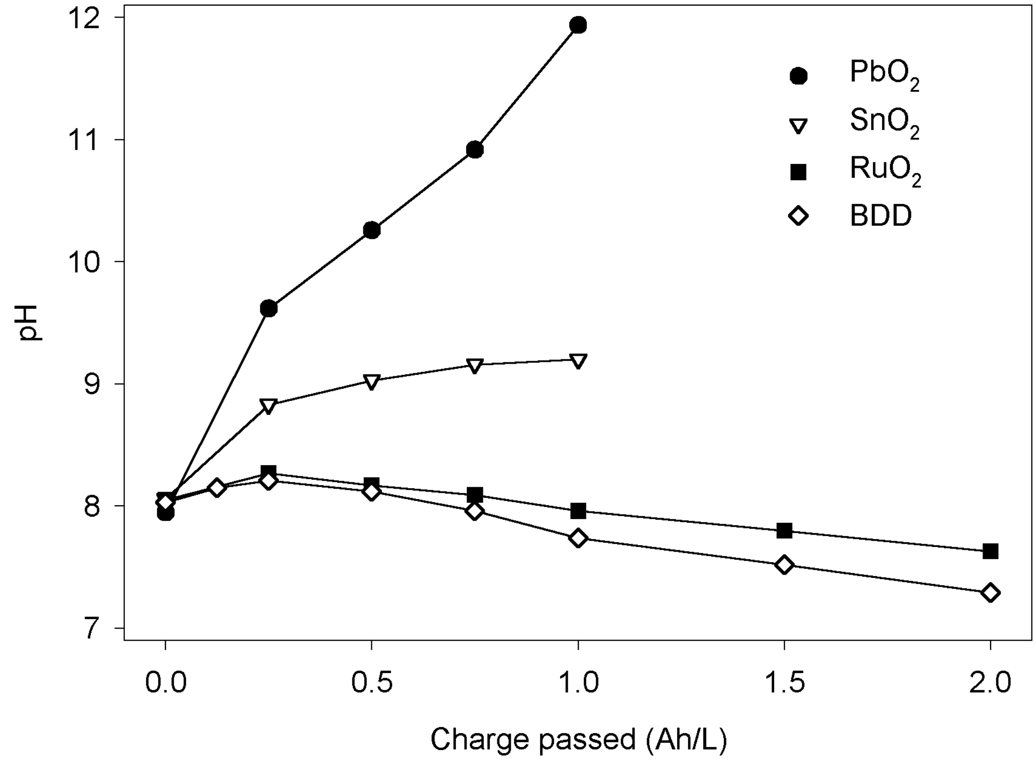

Figure 6 Evolution of the pH during bulk electrolysis of the brine sample 3 at different anode surfaces under galvanostatic conditions (20 mA/cm²)

In Table 2, the changes in a number of routine parameters after the passage of a charge of 1 Ah/L (As in all the experiments 1 L of solution was used, the total input of electrical charge can be expressed in Ah/L) are mentioned. Notably, a similar decrease in absorbency for all electrode materials was measured, while the PbO_2 and SnO_2 electrodes showed similar removal efficiencies for COD and TAN. However, the drastic pH rise when electrolysis was performed with these anodes (Figure 3) resulted in excessive precipitation in the electrolysis cell and on the electrode surfaces. This rendered the electrode surfaces inept with regard to further oxidation and resulted in a steadily increasing cell potential (results not shown). In the case of the PbO_2 electrode, the precipitate contained considerable amounts of Ca^{2+} and Mg^{2+} (58.3 and 3.58 mg/g dry matter respectively), which corroborated with the disappearance of these compounds from the electrolyte. Indeed, it was observed that the soluble Mg^{2+} and Ca^{2+} concentrations decreased more than 90 % (Table 2). Furthermore, solubilisation of the PbO_2 electrode occurred as a concentration of 0.77 mg Pb/L was measured (initial concentration < 0.3 mg Pb/L). Based on these results, no further experiments were carried out on the PbO_2 and SnO_2 anode electrode surfaces.

Table 2 Changes in routine parameters after the passage of a charge of 1 Ah/L during the galvanostatic electrolysis of brine sample 3 at different anodic surfaces. X = no formation or decrease observed

Parameter	Unit	BDD	RuO_2	PbO_2	SnO_2
Active chlorine formation	mg/L	0.73	0.73	X	X
ClO_3^- - Cl increase	mg/L	76.2	10.2	3.77	X
TAN decrease	%	47.7	42.0	13.8	6.5
COD decrease	%	56.2	11.7	18.6	21.7
Absorbency decrease (455 nm)	%	74.1	74.6	74.1	70.3
Ca^{2+} decrease	%	18.3	X	96.8	47.6
Mg^{2+} decrease	%	13.0	X	92.3	36.8

Figures 7.a and b show the COD and TAN removal from brine sample 3 for an applied current density of 10, 20 and 30 mA/cm² for the RuO_2 and BDD anode, respectively. In this range, the current density does not appear to have an influence on the COD or TAN removal efficiency as similar removals are observed for the same charge passed, regardless of the applied current density. In addition, both anodes display a linear removal trend for both COD and TAN removal. From the linear regression curves fitted, a correlation coefficient r^2 of 0.909 and 0.980 is calculated for COD, respectively TAN for the RuO_2 anode; these values equal 0.960 and 0.918 in the case of the BDD electrode. In fact, in all experiments a linear relationship in the removal of TAN and COD is noticed for electrical charges Q < Q* (Ah/L).

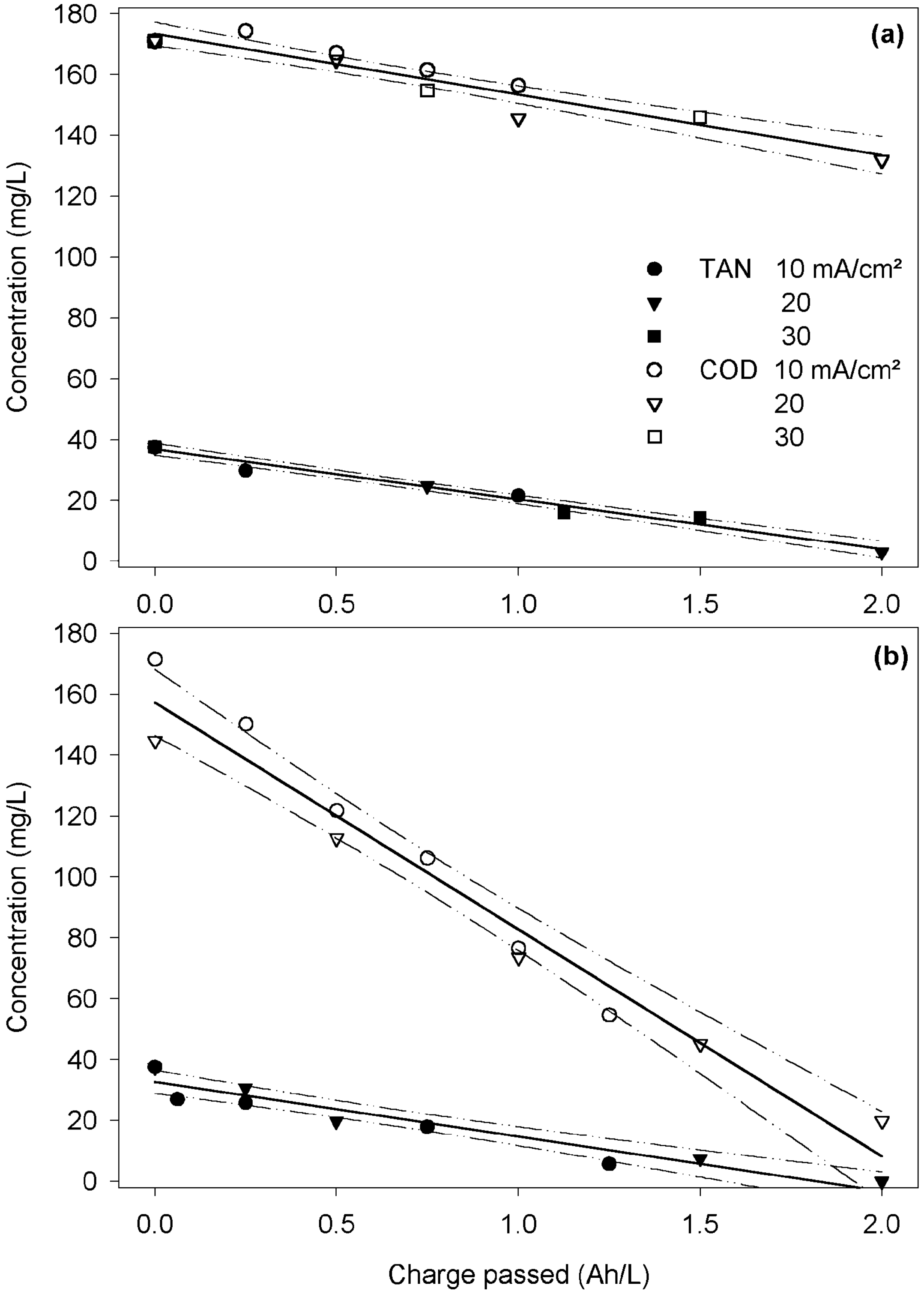

Figure 7 Influence of applied current density on the removal of TAN and COD from brine sample 3 using (a) RuO_2 and (b) BDD. Current densities 10, 20 and 30 mA/cm², galvanostatic conditions. The solid lines indicate the linear regression curve fitted, while the dashed lines show the 95 % confidence interval

The observed values of Q*, as well as the computed COD and TAN removal rates are summarised in Table 3 for the different brine samples. Removal rates determined for the BDD anode are consistently higher than for the RuO_2 electrode, both for COD and TAN, while the values for Q* are lower. Also, the current efficiency (η) and specific energy consumption (E_{sp}) were calculated using the equations from Appendix A. Because higher cell potentials were measured when diamond was used, the specific energy consumption for TAN removal is higher for the latter. On the other hand, the current efficiency is considerably higher for the BDD anode.

Table 3 Comparison of the linear range (Q*, Ah/L) and the computed removal rate (r, mg/Ah), total current efficiency (η_{TOT}, %) and specific energy consumption (E_{sp}, kWh/kg) for the different brine samples and anode surfaces studied. X = not done

Anode	Parameter	Unit	Sample1	Sample 2	Sample 3
BDD	Q*	Ah/L	X	1	1.6
	r_{COD}	mg/Ah	X	114	74.5
	r_{TAN}	mg/Ah	X	22.2	17.9
	η_{TOT}	%	X	50.9	35.2
	$E_{sp, COD}$	kWh/kg COD	X	83.4	188
	$E_{sp, TAN}$	kWh/kg TAN	X	428	782
RuO_2	Q*	Ah/L	1.25	1.5	1.9
	r_{COD}	mg/Ah	44.6	62.9	20.0
	r_{TAN}	mg/Ah	21.9	19.3	13.5
	η_{TOT}	%	27.5	32.2	14.5
	$E_{sp, COD}$	kWh/kg COD	168	119	425
	$E_{sp, TAN}$	kWh/kg TAN	343	388	628

Figure 8 visualises the impact of the initial chloride content of the brine sample on the current efficiency. An increase of the chloride content from 595 to 804 mg/L (brine sample 3 vs. 2) results in an increase of the current efficiency of 45 % for the BDD anode, while for the RuO_2 electrode, the current efficiency roughly doubles.

In Figure 9, the evolution of inorganic chlorinated species during the electrolysis of brine sample 2 is indicated for both anodes. A declining chloride concentration is measured; concomitantly, the chlorate concentration increases. Both trends are more pronounced for the BDD electrode.

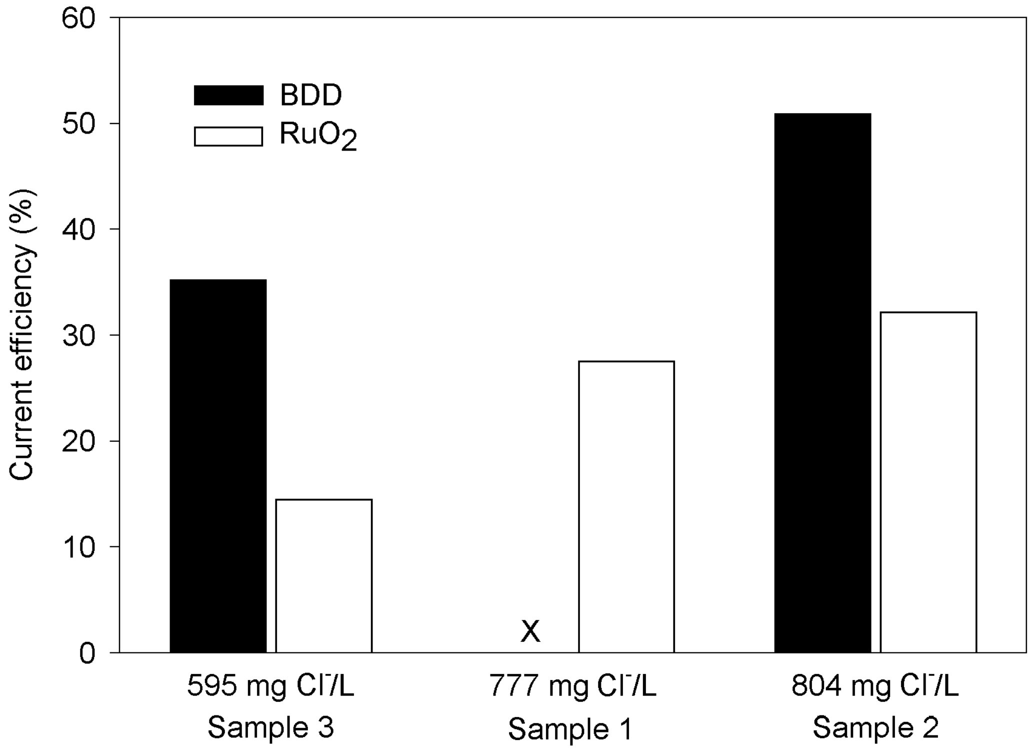

Figure 8 Influence of the chloride content of the brine samples (mg/L) on the current efficiency (%) for RuO_2 and BDD anodes. X = not done

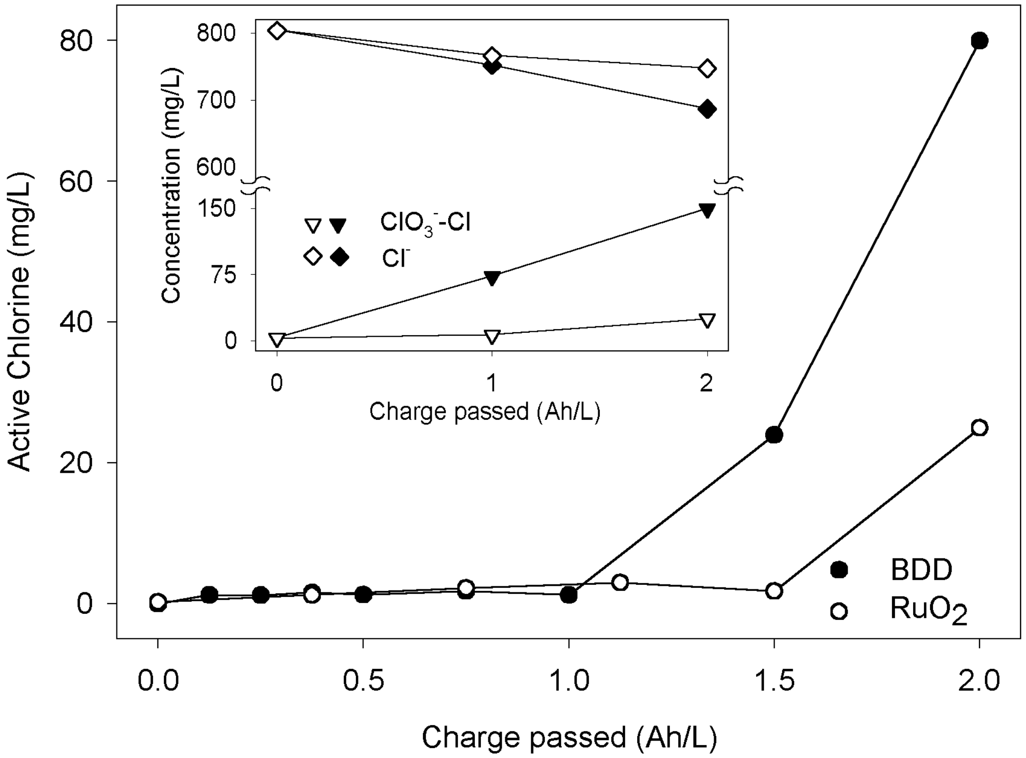

Figure 9 Active chlorine concentration (mg/L) during the electro-oxidation of brine sample 2 at RuO_2 and BDD electrodes. Inset shows the evolution of chloride and chlorate concentration during the experiment

The active chlorine concentrations are lower than 1 mg/L until a passed charge of 1.5 and 1 Ah/L for the RuO_2 and BDD electrode, respectively. Notably, this value corresponds to the observed value of Q* (Table 3). For electrical charges Q > Q*, free active chlorine is evolved and high active chlorine concentrations are measured. From the linear regression curve fitted (not all data are shown in Figure 9), a chlorine evolution rate of 130 mg/Ah and 84 mg/Ah was computed for the BDD and RuO_2 anode, respectively.

More details are revealed in Figure 10 for brine sample 1 using a RuO_2 electrode; however, comparable trends were seen for the other brine samples and the BDD electrode (results not shown). The TAN removal is linear (the correlation coefficient equals 0.959 in the range 0 - 1.25 Ah/L) and the chlorine evolution starts when a substantial amount of TAN is oxidised. For a charge $Q > Q^*$, the evolution of active chlorine content can be linearised ($r^2 = 0.995$) and a production rate of 139 mg/Ah can be calculated. Using the data from Table 3, the average removal at the start of chlorine evolution is assessed to be 81 and 31 % for the RuO_2 anode and 69 and 61 % for the BDD anode, for TAN and COD respectively.

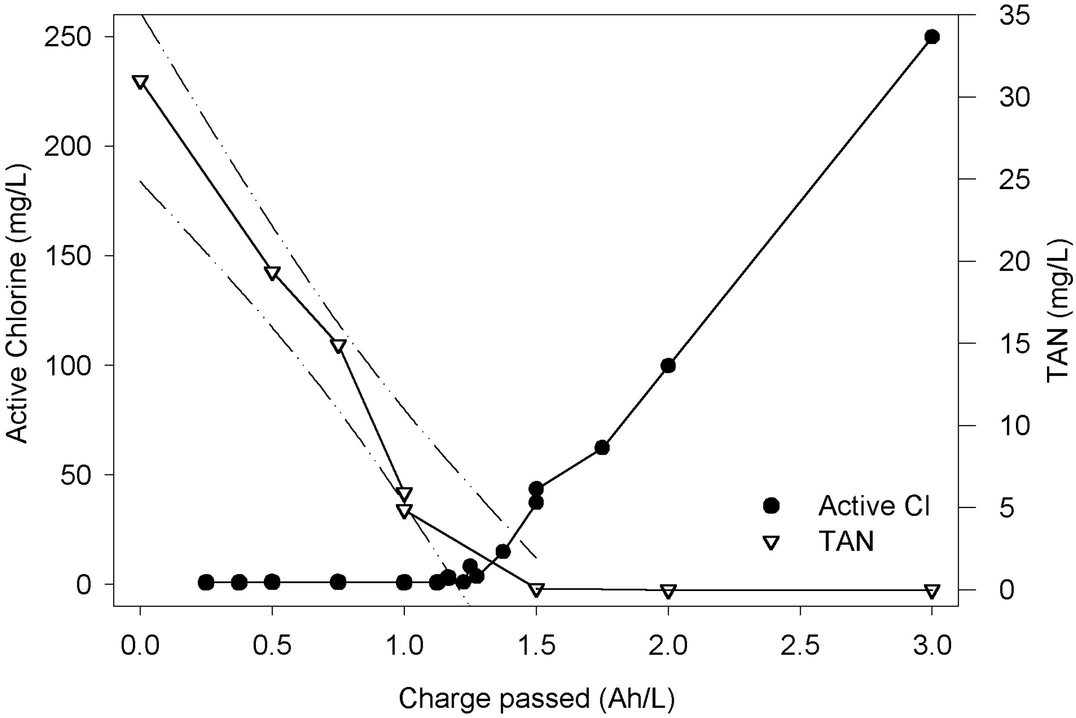

Figure 10 Active chlorine concentration and TAN concentration (mg/L) during the electrolysis of brine sample 1 at a RuO_2 anode. The dashed lines indicate the 95 % confidence interval of the regression curve fitted to the TAN data in the range 0-1.5 Ah/L (Linear regression curve omitted for reasons of clarity)

The percentual decrease in absorbency for five wavelengths during the oxidation of brine sample 3 on RuO_2 and BDD is displayed in Figures 11.a and 11.b, respectively.

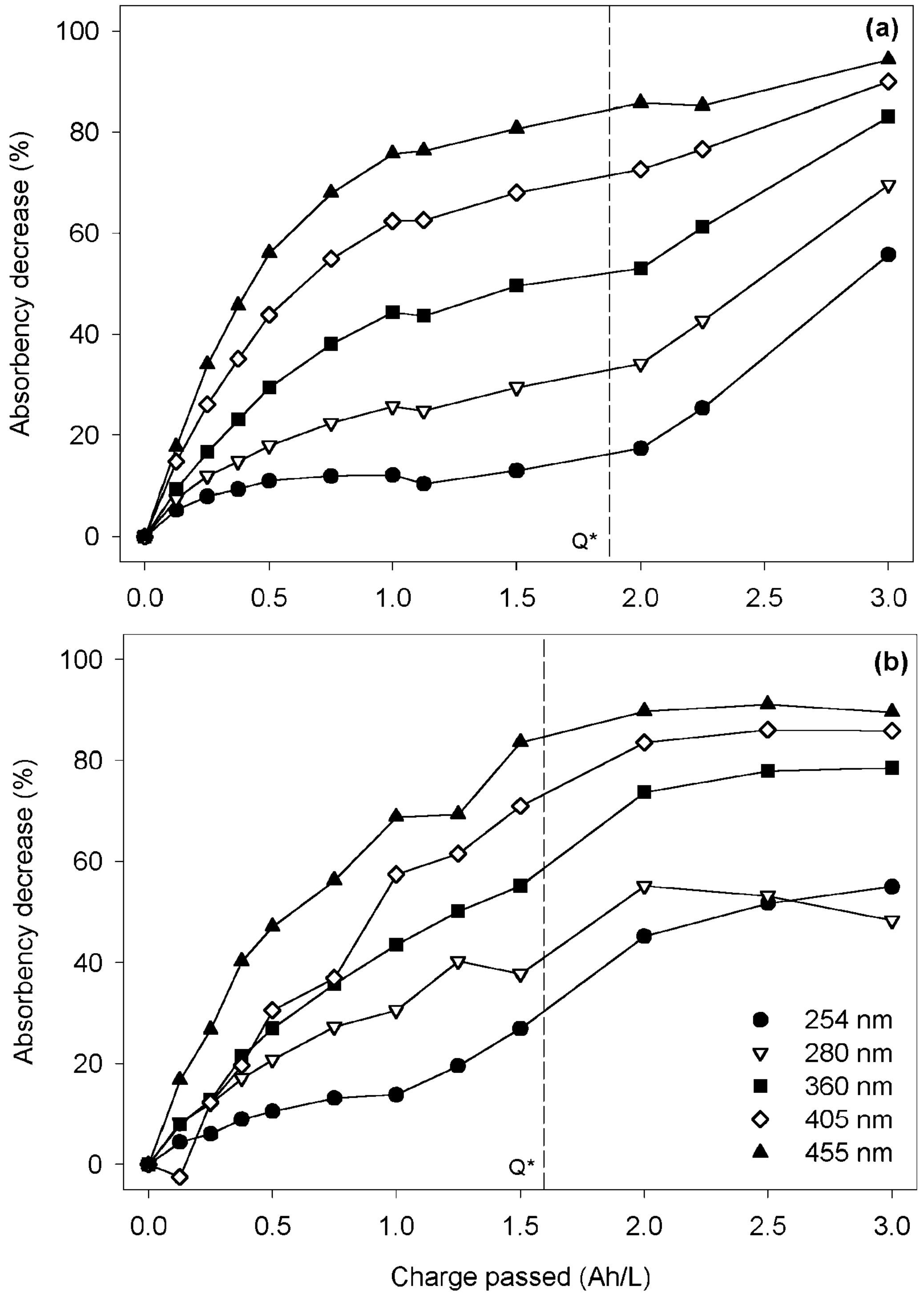

Figure 11 Percentual decrease in absorbency for five wavelengths (254, 280, 360, 405 and 455 nm) during the oxidation of brine sample 3 on the RuO_2 (a) and on the BDD (b) electrode. The dashed line indicates the start of chlorine evolution (charge Q*)

The electrolysis has a decremental effect on the absorbency: for the visible wavelengths (360 - 455 nm), a decrease of more than 80 % is realised, while in the UV range (254 - 280 nm), around 50 % reduction is accomplished for a charge input of 3 Ah/L for both anodes. However, after passage of a charge Q*, the absorbency decrease at low wavelengths is remarkably higher for the BDD electrode. After a charge input of 3 Ah/L the electrolyte exhibits a residual absorbency of 0.76 and 1.35 % (at 455 nm, relative to demineralised water) for the RuO_2 and BDD experiment respectively, illustrating a nearly complete removal of colour.

DISCUSSION

The experiments performed on the BioMAC reactor indicate a very poor biodegradability of the tested reverse osmosis concentrates (Figure 3). Although satisfactory during the first period, the poor COD removal during the second period can be attributed to the nature of the RO concentrate. The latter originated from a mixed textile and domestic wastewater effluent and was characterised by an offensive purple colour. This suggests the presence of a substantial amount of textile dyes that are known to possess a very low biocompatibility (Vandevivere *et al.*, 1998). Although these dyes can be successfully sorbed onto activated carbon (Shen *et al.*, 2001) and the increased salinity of the RO brine was not expected to affect the biological activity of the BAC filter (Sozanski, 1995), the applied adsorptive/bioregenerative BioMAC treatment was not able to achieve substantial removal. On the other hand, nitrification did occur, and from the comparison of in- and output of the reactor nitrogen content it can be deduced that no stripping of ammonia took place (Figure 2). From this, it is concluded that the RO concentrate, though recalcitrant, was not acutely toxic since nitrifying micro-organisms are known to be sensitive towards influent toxicity (Gernaey *et al.*, 1997).

As an alternative approach, the electrochemical oxidation of RO concentrate was considered. First, the electrochemical behaviour of the electrodes in RO brine was studied by cyclic voltammetry. The small anodic current peak observed at 1.6 V vs. SHE during the first polarisation cycle for the BDD electrode can be linked to modification of the electrode surface (Figure 4.a). More specifically, this has been ascribed to the formation of oxygen containing functional groups (diamond oxide), which can act as a barrier for electron transfer (Gandini *et al.*, 2000; Notsu *et al.*, 2000). The latter is consistent with the fact that further anodic polarisation in the potential region supporting electrolyte decomposition did not result in any noticeable electrode reactivation - which would normally be the case for electrode fouling (Panizza *et al.*, 2001a). After the formation of an oxide film, evidenced by the oxidation peak in the first cycle, the Ti electrode displays a relatively inert behaviour as low currents are observed during the second cycle (Figure 4.b). The RuO_2-coated electrode shows a good activity as well as an excellent stability, since the consecutive cyclic voltammograms overlap (Figure 4.c).

During the bulk electrolysis batch experiments, the fate of the pollutants present in RO membrane retentate (exemplified here as COD, TAN and colour) was studied. Because RO typically contains substantial amounts of chloride, electrochemical destruction of the contaminants present can be accomplished both by direct anodic electron transfer and by indirect bulk oxidation through the anodic generation of chlorine/hypochlorite (Chiang *et al.*, 1995). As a result, the overall reaction scheme becomes very complicated. An overview of the most important occurring electrochemical reactions is provided in Table 4 (Chiang *et al.*, 1995; Comninellis and Nerini, 1995; Marincic and Leitz, 1978; Wang *et al.*, 2001).

Table 4 Overview of possible electrochemical reactions during the electrolysis of a complex chloride containing wastewater (Chiang *et al.*, 1995; Comninellis and Nerini, 1995; Marincic and Leitz, 1978; Wang *et al.*, 2001)

	Description	Reaction	
Anode	Chlorine evolution	$2Cl^- \rightarrow Cl_2 + 2e^-$	(1)
	Chlorate formation	$OCl^- + 2\ H_2O \rightarrow ClO_3^- + 4\ H^+ + 4e^-$	(2)
	Oxygen evolution	$2\ H_2O \rightarrow O_2 + 4\ H^+ + 4\ e^-$	(3)
	Direct TAN oxidation	$NH_3 + 3\ OH^- \rightarrow 0.5\ N_2 + 3\ H_2O + 3\ e^-$	(4)
	Direct COD oxidation	$COD \rightarrow \text{intermediate} + z\ e^-$	(5)
		$\rightarrow \text{final product} + z'\ e^-$	
Solution	Chlorine hydrolysis	$Cl_2 + H_2O \rightarrow HClO + Cl^- + H^+$	(6)
	HOCl dissociation	$HClO \rightarrow OCl^- + H^+$	(7)
	Chlorate formation	$ClO^- + 2\ HClO \rightarrow ClO_3^- + 2\ H^+ + 2\ Cl^-$	(8)
	Ammonia hydrolysis*	$NH_3 + H_2O \rightarrow NH_4^+ + OH^-$	(9)
	Bulk TAN oxidation**	$2\ NH_3 + 3\ HClO$	(10)
		$\rightarrow N_2 + 3\ H_2O + 3\ Cl^- + 3\ H^+$	
	Bulk COD oxidation	$COD + y\ OCl^- \rightarrow \text{intermediate'} + y'\ OCl^-$	(11)
		$\rightarrow \text{final product'}$	
Cathode	Hydrogen evolution	$2\ H_2O + 2\ e^- \rightarrow H_2 + 2\ OH^-$	(12)

* $pK_a = 9.25$ ** Through the formation of chloramines and subsequent degradation

For the PbO_2 and SnO_2 anodes used in this study, a pronounced pH increase and subsequent precipitation and electrode scaling was noted during electrolysis (Figure 6). Indeed, cathodic reduction reactions such as hydrogen evolution (eq. 12) typically result in a pH rise, which can lead to the formation of insoluble species such as $CaCO_3$, $Mg(OH)_2$ or $MgNH_4PO_4$ (struvite) (Kraft *et al.*, 1999a). In fact, this phenomenon has been implemented for the electrochemical elimination of such components from water and wastewater (Bannoud *et al.*, 1993; Dellamonica *et al.*, 1980). However, in an undivided electrolytic cell, this pH increase is usually matched by anodic reactions such as oxygen evolution (eq. 3) or chlorine hydrolysis (eq. 6). It is therefore plausible that other anodic reactions, *e.g.* electrode dissolution, took place. The latter is substantiated by the recorded soluble Pb concentration, which indicates low electrode stability. This, in turn, might be due to the electrode preparation technique.

Two stages can be distinguished during the electrochemical treatment of the RO retentate on BDD and RuO_2 anodes. For a charge input lower than Q*, a linear decrease of the TAN and COD concentrations is observed (Figures 7.a and b). Taking into account the pH values measured (Figure 6), it can be calculated that around 95 % of the TAN is present as the hydrolysed NH_4^+-ion (eq. 9). Hence, direct oxidation as well as ammonia volatilisation are negligible since both are distinctly favoured by the presence of undissociated NH_3 (Marincic and Leitz, 1978). Consequently, TAN removal can be ascribed to the indirect oxidation effect of the electrogenerated chlorine/hypochlorite (eq. 10). On the other hand, the higher removal rates for both TAN and COD on the BDD anode seem to be related to the higher chlorine production rate of the latter (Figure 9). This substantiates the hypothesis that the COD is also mainly removed through indirect bulk oxidation in this stage (eq. 11). The very low level of active chlorine is a further indication that TAN and COD promptly react with the electrogenerated oxidants; an observation also made by Bonfatti *et al.* (2000b) and Comninellis and Nerini (1995). Once the electrical charge surpasses an amount Q*, a linear increase in active chlorine content is obtained (Figures 9 and 10). According to Comninellis and Nerini (1995), this indicates that the rate of hypochlorite oxidation of the residual pollutants has become very low. It is said that the chlorine demand of the wastewater has been met and this process is usually termed breakpoint chlorination (Kraft *et al.*, 1999b). Beyond this point, further oxidation probably involves electrochemical surface mediated reactions (Bonfatti *et al.*, 2000b).

When comparing both the anode materials, considerable kinetic differences emerge. First, the BDD electrodes are more efficient in the production of active chlorine, as higher rates of active chlorine production were detected. Moreover, higher amounts of chlorate were detected during electrolysis when using BDD electrodes (Figure 9). Because the rate of chlorate formation through bulk reaction with hypochlorite according eq. 8 has been observed to be extremely low (Kraft *et al.*, 1999b), it can be concluded that also the electrogeneration of chlorate is more pronounced at BDD anodes (eq. 2). This excellent performance is confirmed by the work of Ferro *et al.* (2000), who linked the superior chlorine production rate of this material to the high selectivity towards the chlorine evolution reaction (*i.e.* less important parasitic oxygen evolution reaction). Taking into account a predominantly indirect oxidation mechanism, it also justifies the higher removal rates for COD and TAN (Table 3). Second, the extent of destruction prior to hypochlorite formation is substantially higher for the BDD electrode. The latter has been connected to the weak adsorption properties due to the inert surface of the BDD material, resulting in a lower stability and consequently a higher reactivity of chloro- and oxychloro-radicals coadsorbed at the electrode surface (Bonfatti *et al.*, 2000b; Ferro *et al.*, 2000). Hence, the BDD electrode is not only superior with regard to chlorine production, but also with respect to the generation of reactive radicals.

Although the RO brine was characterised by an offensive colour, a complete decolouration is achieved for both anodic materials (Figures 11.a and b), supporting the fact that electrolysis is highly suited for pigment removal. For example, Vlyssides *et al.* (2000) also noticed complete disappearance of colour in a textile dye wastewater using a Ti anode, while Shen *et al.* (2001) successfully degraded 29 textile dyes in an electrochemical activated carbon fibre reactor. However, Panizza *et al.* (2000a) noted that colour removal was unsatisfactory when no chloride was present, thus validating the importance of electrogenerated hypochlorite in the process. This mechanism is authenticated in the present work, since around 80 % of the colour is removed during the first phase of the oxidation. The absorbency at low wavelengths (*i.e.* 280 nm) has been used as an assessment of the refractory organic pollutants present (Seo and Ohgaki, 2001). In this respect, it is shown that the applied electrochemical treatment is capable of removing more than 50 % of these constituents. Again, the higher activity of the BDD electrode is illustrated by a higher removal at equal charges passed.

In the anodic incineration of pollutants, the current efficiency is used as a parameter describing the overall energy-efficiency of the process. In this respect, the influence of the chloride ion has been extensively documented (Bonfatti *et al.*, 2000a; Chiang *et al.*, 1995; Comninellis and Nerini, 1995; Iniesta *et al.*, 2001a; Panizza *et al.*, 2000a). These authors found hat the addition of significant amounts of NaCl (in the order of grams per litre) increased the current efficiency tremendously. An analogous trend is seen for the different brine samples under study, although the samples of the present study displayed lower chloride concentration levels (Figure 8). For a comparable chloride content, the RuO_2 anode displays a similar current efficiency as the Pt electrode system tested by Panizza *et al.* (2000a) in his study of a wastewater containing polyaromatic pollutants (32 % and 25 %, respectively), while the BDD anode outperforms both. In comparison with other studies, the computed specific energy consumption E_{sp} is rather high as values of 12 - 30 kWh/kg COD for the combustion of phenol on boron-doped diamond electrodes (Panizza *et al.*, 2001b) and 91 kWh/kg COD for landfill leachate oxidation on Ti/SnO_2 anodes (Wang *et al.*, 2001) have been reported. However, in these cases highly conductive electrolytes were under investigation, resulting in a rather low cell potential and consequently a lower value of E_{sp} (eq. A.9 and A.10).

CONCLUSIONS

Because of the recalcitrance of the RO concentrate, no satisfactory COD removal could be obtained during biological treatment using the BioMAC reactor. In order to achieve satisfactory biological breakdown, pre-treatment of the RO concentrate with a powerful oxidant (H_2O_2, ozone) and subsequent biological mineralisation of the intermediates should be taken into consideration.

Alternatively, electrochemical oxidation seems a very promising treatment technology: from the batch electrolysis experiments performed in this study, it is clear that the pollutants present in reverse osmosis membrane concentrates (TAN, COD, and colour) can be successfully eliminated using the electrochemical approach. The following observations were made:

1. SnO_2 and PbO_2 anodes show low stability and give rise to high pH values and subsequent precipitation of calcium and magnesium compounds.
2. Concerning the RuO_2 and BDD anodes, the following phenomena are observed:
    - Oxidation is predominantly achieved through indirect hypochlorite bulk oxidation; the higher oxidation rate and extent for the BDD anode can be assigned to the higher selectivity for chlorine evolution and the higher reactivity of co-adsorbed radicals.

- Overall performance of the BDD electrode was higher than for RuO_2: higher rates for TAN and COD removal, better current efficiencies and a faster removal of colour were noted.

Hence, especially the BDD anode merits additional research with regard to further elucidation of the oxidation mechanism and the possible formation of organo-chlorinated components.

ACKNOWLEDGEMENTS

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APPENDIX A: LINEAR REGRESSION MODEL FOR CONCOMITANT COD AND TAN REMOVAL

The linear equation predicting the TAN and COD concentration as a function of the charge Q (Ah/L) passed through the electrolysis cell can be written as:

$$(COD)_Q = (COD)_0 - r_{COD} Q \qquad (A.1)$$

$$(TAN)_Q = (TAN)_0 - r_{TAN} Q \qquad (A.2)$$

where $(COD)_0$ and $(TAN)_0$ are the initial COD and TAN concentrations (g/L) and r is the removal rate (g/Ah). The instantaneous current efficiency (ICE) for COD removal is defined as (Comninellis and Pulgarin, 1991):

$$ICE_{COD} = 4FV \frac{\left[(COD)_t \quad (COD)_{t+\Delta t}\right]}{I\,\Delta t} \frac{1}{32} \qquad (A.3)$$

where $(COD)_t$ and $(COD)_{t+\Delta t}$ are the CODs at time t and t+Δt (g O_2/L) respectively, I is the current (A), F is the Faraday constant (96487 C mol^{-1}), V is the volume of the electrolyte (L), 4 is the number of exchanged electrons per mole of O_2 consumed and 32 is the molecular mass of oxygen (g O_2/mol). This equation can be rewritten as:

$$ICE_{COD} = \frac{F}{28800}\frac{\left[(COD)_{Q} \quad (COD)_{Q+\Delta Q}\right]}{\Delta Q} \tag{A.4}$$

where $[(COD)_Q\text{-}(COD)_{Q+\Delta Q}]$ is the decrease in COD (g O_2 /L) after the passage of a charge per reactor volume ΔQ (Ah/L). Since the electrochemical oxidation of ammonia requires the exchange of 3 electrons (Table 4), an analogous equation can be written for the ICE for TAN removal:

$$ICE_{TAN} = \frac{3F}{14 * 3600}\frac{\left[(TAN)_{Q} \quad (TAN)_{Q+\Delta Q}\right]}{\Delta Q} \tag{A.5}$$

with 14 the molar mass of TAN (mg N/mol). Substitution of eq. (A.1) in (A.4) and eq. (A.2) in (A.5) respectively yields

$$ICE_{COD} = \frac{F}{28800} r_{COD} \tag{A.6}$$

$$ICE_{TAN} = \frac{F}{16800} r_{TAN} \tag{A.7}$$

Because the removal is linear, eq. (A.6) and (A.7) are constants. Hence, the ICE equals the average current efficiency η for both TAN and COD. The overall current efficiency is calculated as the sum of both the TAN and COD current efficiency:

$$\eta_{TOT} = \eta_{COD} + \eta_{TAN} \tag{A.8}$$

The specific energy consumption (E_{sp}), expressed as kWh/kg COD or TAN removed, is directly related to the removal rate since the latter is written as g COD or g TAN removed / Ah:

$$E_{sp,COD} = \frac{V_c}{r_{COD}} \tag{A.9}$$

$$E_{sp,TAN} = \frac{V_c}{r_{TAN}} \tag{A.10}$$

Where V_c is the cell potential (V).

GENERAL DISCUSSION AND FUTURE PERSPECTIVES

CURRENT STATUS OF WASTEWATER RECLAMATION, REUSE AND RECYCLING

Water is the key element of life, but it is also the most undervalued commodity known to humanity. Especially in the developed countries, water is taken for granted and an endless flow of pure, clean water is expected by simply opening the tap. Yet water is becoming increasingly scarce, even in countries thought to have an affluent water supply. As discussed in Chapter 1, Belgium is a prime example of the latter, despite its moderate sea climate and 821 mm of annual precipitation (KMI, 2002). The increasing stress on the existing water reserves, coupled to water quality issues, is prompting government and industry to seek out and explore alternative water resources. This has sparked a heightened interest in the concepts of water reuse, reclamation and recycling.

The reclamation and recycling of wastewater has an enormous potential: by creating shortened loops in the natural water cycle, a seemingly inexhaustible supply of "new" water can be obtained. Effluent originating from centralised WWTPs is, because of its availability, quality and quantity, exceptionally suited for a wide range of reuse applications (Asano, 1994). However, to avoid adverse effects and to protect public health, the quality of reclaimed wastewater has to be explicitly guaranteed (Dewettinck, 2002). In addition to rigorous monitoring, in most cases treatment prior to reuse is necessary. Therefore, emphasis is placed on the importance of efficient, reliable, sustainable and cost-efficient advanced wastewater reclamation technologies.

Technological solutions

The implementation of pressure driven membrane processes, such as MF, UF, NF, RO and their hybrid derivatives has become the cornerstone of advanced wastewater treatment and reclamation systems. Membrane technology has reached full stature and many full-scale installations in drinking water production and wastewater reclamation are in operation throughout the world; many more are planned (Bonnélye *et al.*, 2001; Côté *et al.*, 2001; Gander *et al.*, 2000a; Gnirss, 2000).

Treating WWTP effluent to a quality suitable for unrestricted non-potable reuse can be readily achieved by hybrid MF or UF processes. In Chapter 5 of the current work, this was demonstrated for a combined BAC-MF reactor. The tested BioMAC reactor had a polishing effect on the WWTP effluent, as feed concentration fluctuations were substantially attenuated and odorous constituents significantly removed. The permeate of BioMAC reactor could comply with the US EPA regulations for unrestricted non-potable reuse, provided (i) a periodic replacement of the AC filter was carried out and (ii) a post-treatment chlorination was performed.

Higher permeate quality, especially with respect to the removal of medium and low molecular weight organic components and dissolved solids, typically requires the implementation of NF (Irvine *et al.*, 2001) or RO (Downing *et al.*, 2002). As an alternative to filtrative desalination, evaporation was considered in Chapter 4 of this thesis. The process allows for a similar (or even superior) decontamination of WWTP effluent as RO desalination, although the abatement of volatile components necessitated a Fenton oxidation pre-treatment. From an engineering point of view, evaporation is an elegant, simple and robust process, but the required energy input is relatively high. Currently, the energy expenditure for RO is in the range of 3 kWh/m^3 (Buros, 2000), while evaporative treatment typically requires 6 - 8 kWh/m^3 (Wade, 2001). In Table 1, a cost comparison is made between the evaporative process (Chapter 4), the BioMAC process (Chapter 5) and a RO process for a plant producing 100 m^3/h treated water. Utmost care must be taken upon comparison of these costs, as calculation methods may differ substantially for data originating from different sources. However, the calculated values can serve as a rough guideline.

Table 1 Capital and operational expenses (EUR/m^3 produced water) for Fenton-mediated evaporation, BioMAC and RO filtration. Data for the RO plant obtained from Van Houtte *et al.* (1999) and Filteau *et al.* (1995)

	Evaporation	**BioMAC**	**RO**
Capital expenses [a]	0.43	0.15	0.40
Operational expenses, of which:	1.18	0.38	0.26
Energy requirement [b]	*0.40*	*0.05*	*0.15*
Reagents [c]	*0.78*	*0.33*	*0.11*
TOTAL	**1.61**	**0.53**	**0.66**

[a] Interest rate 7 %, depreciation over 15 year, maintenance is 3 % of investment per year
[b] 0.05 EUR/kWh
[c] $FeSO_4$, H_2O_2 and H_2SO_4 for the evaporative process and GAC for the BioMAC reactor

From these data it is apparent that the proposed membrane processes offer a more cost-efficient solution as both the predicted BioMAC and RO treatment cost are substantially lower than for an evaporation based treatment train. Clearly, the high energy requirements restrict the use of evaporation as a wastewater reclamation technology to sites where an excess amount of steam or electricity is available. For example, Rose *et al.* (1999) proposed the integration of evaporation in a combined municipal solid waste, wastewater treatment and reclamation process. In the scheme, visualised in Figure 1, low pressure steam and waste heat of the waste-to-energy incineration facility is used to pre-heat, boil and evaporate reclaimed water for the distillation of ultra-pure water. Because of its cyclic character, such an innovative integrated concept embodies the concept of sustainable development and this type of integration may be viewed as a natural and synergistic evolution for many communities.

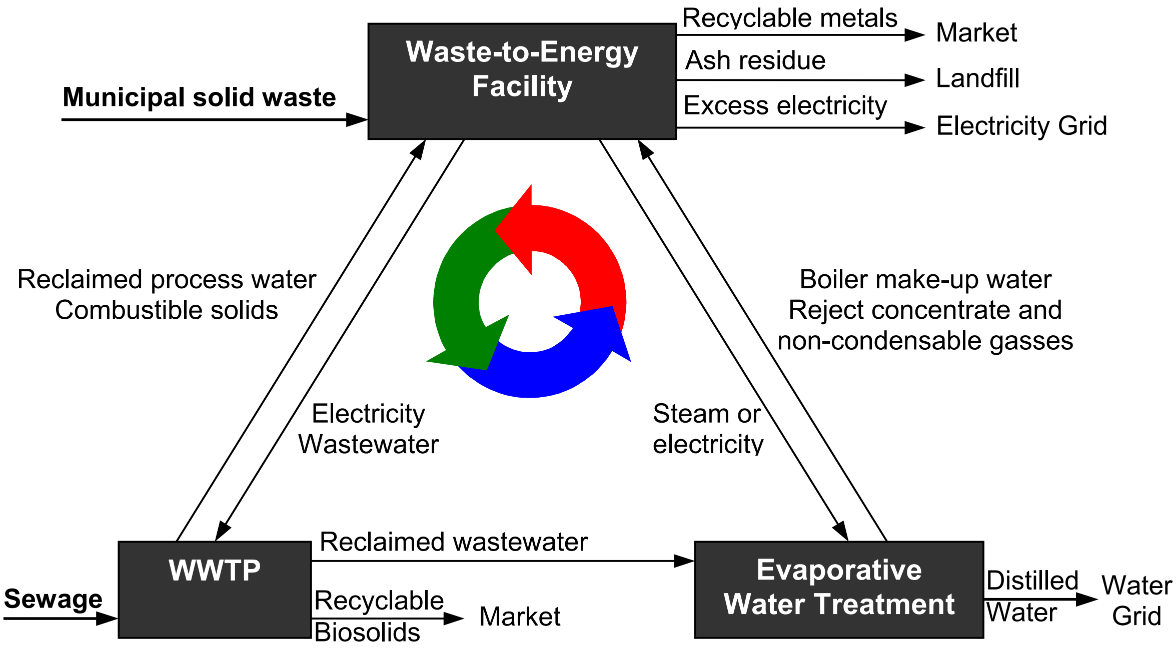

Figure 1 Conceptual diagram for a wastewater treatment plant co-located on a waste-to-energy site (Rose *et al.*, 1999)

However, when taking into account the economic considerations presented in Table 1, it becomes clear that membrane technology must be regarded as the current state of the art technology for many wastewater reclamation and reuse applications. Hybrid processes are increasingly considered for the production of various qualities of water (Fane, 1996; Watanabe *et al.*, 2000), while RO filtration is a more economically viable option for the production of high quality reclaimed wastewater.

Perhaps the most eminent illustration of the immense success of RO as a high quality water treatment process is the operational RO desalination capacity, which increased from an estimated 5 million m^3/day in 1994 (Fane, 1996) to 8.1 million m^3/day in 1998 (Buros, 2000). Informal projections indicate this figure rising to over 20 million m^3/day by 2010 (Mcmann, 2002) However, this has created a new problem as the amount of membrane concentrate has increased proportionally: assuming a process recovery of 90 %, desalination applications alone generated 810000 m^3 concentrate/day in 1998. The disposal of these retentates is being restricted due to increased environmental awareness and more stringent regulations. Indeed, the high dissolved solids content often precludes discharge into freshwater bodies, while disposal to the headworks of a WWTP may cause potential adverse effects on the sewage treatment works (Squire, 2000). According to Ahmed *et al.* (2001), the disposal or treatment of membrane concentrate represents 5 to 33 % of the total desalination price, which implies a cost of 0.33 to 2.2 EUR/m^3 of concentrate treated or disposed .

This problem - and the possible costs associated to it - instigated the research carried out in Chapter 6. Electrolysis was investigated as a novel treatment approach for RO concentrates generated from the filtrative treatment of a mixed domestic and textile wastewater. It was proven that inorganic TAN was completely oxidised to N_2, while organic contaminants (COD, colour) could be successfully eliminated when using boron-doped diamond electrodes, a promising wastewater treatment electrode material. Removal of total dissolved solids - through precipitation of Ca-containing species - was observed on PbO_2 and SnO_2 electrodes. From the data presented in Chapter 6, it can be computed that the BDD electrodes require 33 - 60 kWh/m^3 to completely abate the COD and TAN present in the RO concentrates under scrutiny. This results in a projected energetic cost of 1.65 - 3.0 EUR/m^3 concentrate, which is in the same range as the current costs associated with concentrate disposal or treatment. However, this cost can be significantly reduced by further refining and optimising the design of the electrochemical cell. As a result, the proposed electrochemical process has the potential to become - especially in those cases where high costs are associated with concentrate treatment - a cost-comparable technology.

Impact of emerging pollutants

As discussed in Chapter 2, the field of advanced wastewater treatment and reclamation is being challenged by the continual emergence of new SOCs. The elimination of these hardly eliminable and hardly biodegradable components that seem to possess toxicological effects at very low concentrations has severe technological implications.

- Many pharmaceutical compounds resist primary and secondary treatment and are thus found at µg/L-levels in the effluents of WWTPs, from where they can find their way into surface water and groundwater (Heberer *et al.*, 2001). Therefore, efficient removal of these components during advanced treatment is essential prior to reuse. SAT efficiently eliminates anti-inflammatory and lipid-regulating drugs, but anti-epileptics persist during groundwater recharge, even after a subsurface travel time of more than 6 years (Drewes *et al.*, 2002). Sacher *et al.* (2001) reported that multi-barrier physico-chemical advanced treatment (*i.e.* bankfiltration followed by ozonation and GAC filtration) successfully removed drug residues found in surface water, whereas a single treatment step was insufficient to achieve complete elimination of all monitored pharmaceuticals. Other advanced treatments such as MF or MBR have been found to be inefficient for the more polar pharmaceutical compounds (Pinkston and Sedlak, 2001; Schröder, 2001). So far, the data reported in literature point out that only NF and RO could guarantee the rejection of all drugs to levels below the detection limits (Drewes *et al.*, 2002; Pinkston and Sedlak, 2001).
- Endocrine disruptors can be removed from aqueous streams by physico-chemical processes and the US EPA recommends the use of GAC filtration as the best available technique (US EPA, 2001). Indeed, from lab-scale experimental results, Tanghe (1999) calculated that existing full-scale drinking water GAC filters could successfully remove nonylphenol from fresh water resources. Additional batch experiments showed that AOPs such as Fenton oxidation and ozonation very efficiently removed octylphenol at modest oxidant concentrations. However, the efficiency of these processes can be tremendously reduced when treating wastewater containing interfering species such as humic substances and other organic/inorganic pollutants. MF and UF can retain substantial amounts of endocrine disrupting components, although this is due to the retention of particulate matter, which binds large quantities of the more liphophilic target compounds (Huang and Sedlak, 2001; Witgens *et al.*, 2001). Only NF and especially RO membranes seem to be able to retain - partly by sorption onto the membrane - more than 90 % of the challenged estrogens, even when organic matrix compounds such as fulvic acids or natural organic matter are present (Nghiem *et al.*, 2001).

- Since NDMA seems to be formed during the chlorination / chloramination of (purified) wastewater as a possible DBP, research efforts are focusing on preventing or reducing its formation. Effective control measures include: removal of ammonium prior to chlorination, breakpoint chlorination and avoidance of the use of monochloramine for wastewater disinfection. Once formed, NDMA may be eliminated by ultraviolet irradiation (Mitch and Sedlak, 2002).

The emergence of new pollutants emphasises the absolute importance of advanced treatment in wastewater reuse and reclamation. In order to avoid possible adverse ecotoxicological impacts or deleterious human health effects - or just as a cautionary principle - potable applications of WWTP effluent necessitates the removal of endocrine disruptors and pharmaceuticals to extremely low levels. This can only be achieved by a performant and well-designed multi-barrier treatment approach. Because of its ability to reliably and very efficiently reject these components, membrane filtration by NF or RO should unconditionally be considered an integral part of such a treatment.

FUTURE PERSPECTIVES OF WASTEWATER RECLAMATION

The driving forces for water wastewater reclamation, reuse and recycling are now well-known (Reith and Birkenhead, 1998):

- Shortage of fresh water is one of the main threats to sustainable economic development in the more developed countries - no water means no growth.
- Available water resources are under strain due to pollution, over-extraction and saline intrusion.
- Consumers demand higher water quality and the right to an uninterrupted water supply.
- Tough environmental legislation and discharge levying are raising the cost of water and effluent. Further treatment and reuse, instead of discharge, therefore becomes more attractive.
- Climatic changes may make these problems worse as previously water-rich regions suffer extended droughts.

Economic growth and development, an increasing standard of living and population growth will inevitably mean that - despite conservation measures - more water is needed, especially in urban areas. It is therefore apparent that the concepts of wastewater reclamation, reuse and recycling will become increasingly vital within the framework of sustainable water management.

Role of WWTPs

If the sewerage system is to be considered a source of water for the reclaimed water system, *i.e.* a product to be sold instead of a waste to be discarded, it will require special attention. The wastewaters discharged into the sewerage system must be characterised to assure that troublesome wastewaters do not reach the WWTP. Especially industrial wastewater streams may prove to be toxic to the micro-organisms necessary for the biological wastewater treatment preceding reclamation and therefore require pre-treatment. Infiltration of seawater into sewers or saline wastewater discharges from water softeners could make the reclaimed water unfit for irrigation. In addition to sewage characterisation, rigorous and continuous effluent quality monitoring should be implemented (Figure 2).

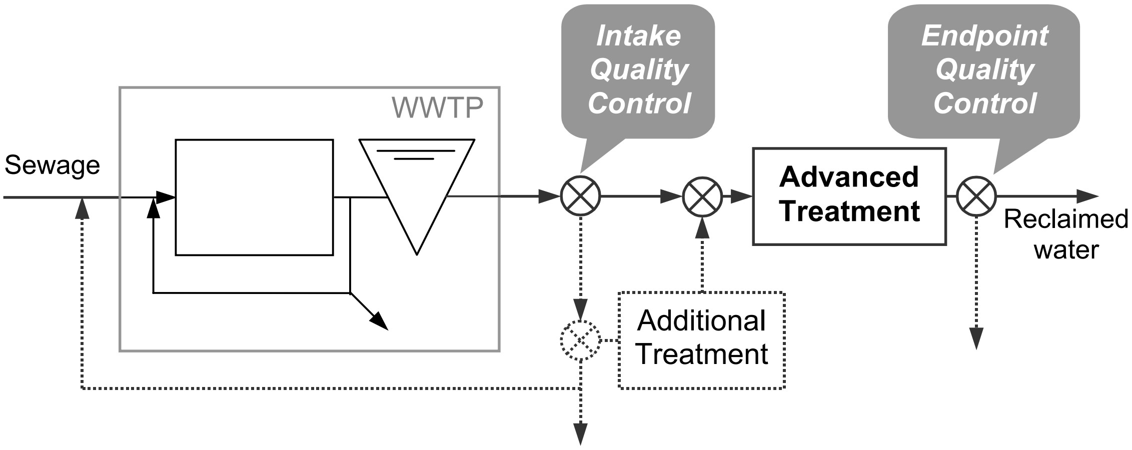

Figure 2 Role of WWTP effluent intake quality monitoring and reclaimed water endpoint quality control (Dewettinck *et al.*, 2001a)

Hygienic and chemical sensors can be applied to rapidly detect effluent of deviant quality and to establish corrective actions, such as additional treatment, re-treatment or discharge. For example, Dewettinck *et al.* (2001a) developed a rapid pH-based biosensor able to detect inefficiently treated effluent within 1 hour. Likewise, an abnormal shift in the amount and/or composition of the volatile components in WWTP effluent could be efficiently detected with an electronic nose (Dewettinck *et al.*, 2001b). The use of such sensors can be extended to monitor the process performance of the applied advanced treatment train and to assess the quality of the final reclaimed water.

If the WWTP is situated at a considerable distance from the existing or new reclaimed water markets, the construction of a so-called satellite reclamation plant, as shown in Figure 3.a, may be appropriate (Okun, 2000). The process has also been termed “water mining”.

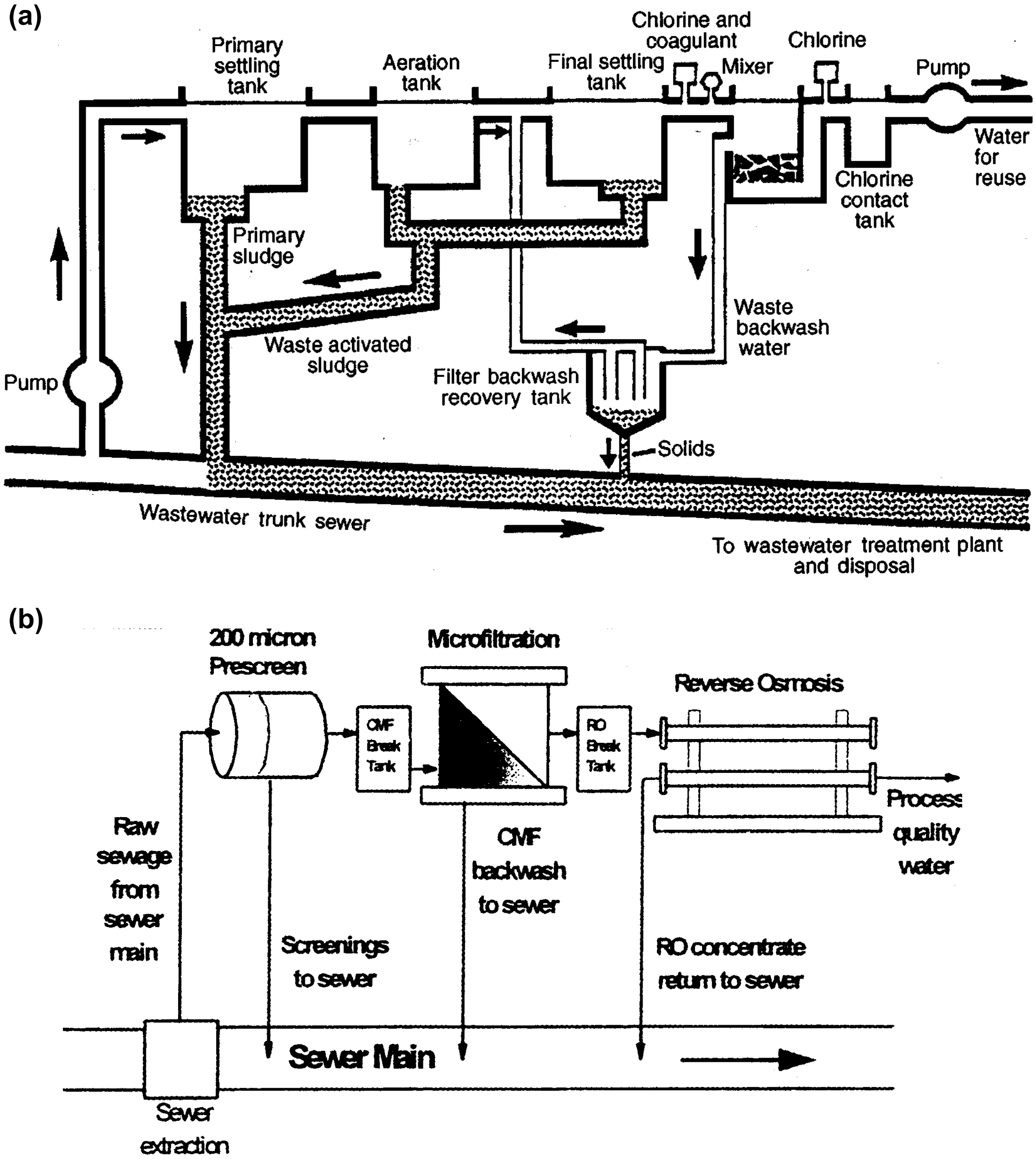

Figure 3 Concept of water mining using satellite wastewater reclamation facilities: (a) satellite activated sludge plant (Okun, 2000) (b) pre-screen / MF / RO treatment train (Johnson *et al.*, 1997)

Such a plant could be considerably smaller because the sludge products are discharged back into the trunk sewer for treatment at the existing treatment plant. As a result, noise and odour emissions are non-existent and a satellite plant can be completely enclosed and even located in a residential or commercial area. Johnson *et al.* (1997) pilot-tested a satellite wastewater reclamation plant consisting solely of membranes. The concept, depicted in Figure 3.b, has proven to be economically and technically feasible and has some additional benefits:

- *Compactness*: a water mining plant processing 2000 m³/day could have a footprint of 400 m^2.
- *Automation*: MF and RO are highly automated, reducing operator need and making the process well suited for telemetric operation and monitoring.
- *Suitability for intermittent operation*: the process can be shut down and readily restarted when required with no loss in treated water quality.

Role of membrane technology

Membrane technology is currently perceived as the state of the art technique for many water and wastewater applications. The main benefits associated with membrane processes include compact footprint, superior effluent quality, reduced sludge production, amenability to automation,.... As the understanding of the fundamentals of membrane processes grows, current and future membrane technology developments can be expected in the following areas:

- Novel methods to alleviate or *prevent fouling and biofouling* (Fane *et al.*, 2000).
- Development of *novel membrane materials*, which are less prone to fouling or require less strict operation conditions. For example, Thin film composite RO membranes operate at half the pressure of cellulose acetate (Reith and Birkenhead, 1998).
- *Optimisation of pre-treatments*, *e.g.* MF can reduce the operational costs of subsequent RO by 70 % (Vedavyasan, 2000).
- *Membrane cost*. Currently, membrane prices are dropping an estimated 12 % annually (Vedavyasan, 2000).
- Development of *large, cost-efficient membrane modules* allowing huge-scale process implementation (Bonnélye *et al.*, 2001; Côté *et al.*, 2001; van der Roest *et al.*, 2001).

Consequently, projections for the next decade envision a tremendous increase of membrane based technology. Vedavyasan (2000) notes and annual increase of 28 % in the application of UF/RO, while (Benedek, 2001) estimated that the amount of wastewater and potable water treated by membranes tripled in the period 1999 - 2001. The predicted market share of membrane technology in water treatment - a 330 billion

EUR market in 2001 - as well as the percentual share in different types of water treatment plants is summarised in Figure 4.

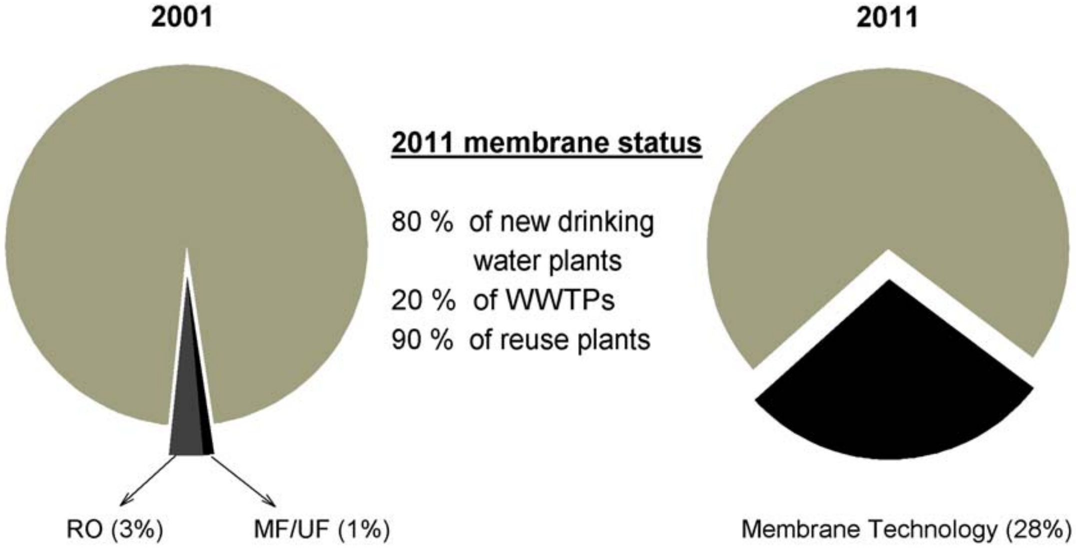

Figure 4 Projected membrane technology status and water technology market share in 2011 (Benedek, 2001)

Towards concentrate treatment technology ?

Taking into account the expected increase of wastewater reuse, the projected predominance of membrane technology, a new issue emerges: treatment of the produced membrane concentrates. Indeed, membrane processes are highly performant as a separation technique, but pollutants are retained and concentrated in the reject brine. For NF and RO, most organic constituents as well as a significant amount of the dissolved solids are found back in the concentrate. Recent literature data suggest a similar fate for emerging pollutants, such as pharmaceuticals and endocrine disruptors. Therefore, disposal of these concentrates should, within the framework of sustainable development, not be considered. Because of its capricious nature, biotechnological treatment of membrane concentrates may prove to be non-viable or should at least be lined up with strong chemical oxidation. Physico-chemical AOPs, such as ozonation, Fenton’s reagent and, as shown in Chapter 6, electrochemical treatment should be explored. The appearance of novel electrode materials that can operate at high current efficiencies, is especially promising in this respect. Because of the presence of chloride in membrane concentrates, attention should be given to the formation of possible DBPs (*e.g.* NDMA) during electrochemical treatment.

Current and projected developments in water reclamation and membrane technology point out that membrane concentrate could become a major bottleneck. Hence, the development of a suitable and sustainable concentrate abatement technology will most likely be primordial to further successful implementation of membrane technology in water and wastewater reclamation.

SUMMARY

Water is the most essential and most precious resource. However, strong indications exist that this crucial asset is becoming increasingly scarce, even in countries thought to have an affluent water supply. Belgium, for example, has a relatively limited supply of renewable water resources and is currently using over 40 % of them. Human activities, such as land sealing, damming, groundwater extraction and diffuse and point source pollution, have a profound impact on the quality and quantity of available water resources. It has therefore become clear that sustainable development of our water supplies will require the careful balancing of demand and supply through integrated water management instead of merely augmenting the supply. Because in many areas conservation measures have been fully exploited, government and industry are exploring alternative water resources. Especially in urban areas, the only substantial “new” resource is the wastewater generated locally.

The concepts of wastewater reclamation, reuse and recycling have an enormous potential: by creating shortened loops in the natural water cycle, a seemingly inexhaustible supply of “new” water can be obtained. A source that is readily available is the effluent originating from centralised wastewater treatment plants (WWTPs). In theory, wastewater can be reused for any purpose, only the level of treatment required to bring it to a quality appropriate for a certain need varies. The potential applications of reclaimed wastewater can be divided in seven categories: agricultural and landscape irrigation, industrial applications, groundwater recharge, environmental / ecosystem restoration, non-potable reuses and direct or indirect potable reuse. WWTP effluent could be used as such in a number of applications, *e.g.* for irrigational purposes, but often requires additional treatment prior to reuse. Indeed, some of the microbiological, chemical and organoleptic constituents must be removed to prevent adverse effects (*i.e.* corrosion, fouling, clogging) and, more importantly, to protect public health. Hence, existing guidelines generally presuppose different reclaimed quality criteria for different applications.

Uses such as non-potable urban reuse or indirect potable reuse generally require some form of tertiary or advanced treatment, which can even be specified in the guidelines. An immense array of advanced treatment technologies exists and it was the scope of the current work to identify and explore suitable technologies for the reuse of WWTP effluent. Emphasis was placed on membrane technology, evaporation and electrolysis.

A first experimental study evaluated the suitability of evaporation as an alternative to filtrative effluent treatment. It was shown that evaporation allowed for extensive decontamination of wastewater treatment plant effluent, as most inorganic and organic contaminants were completely removed. TAN could be retained in the concentrate when pH was adjusted to values lower than 4; however, this led to increased nitrite volatility due to the formation of nitric oxide. Using an electronic nose, it was observed that evaporation gave rise to a change in odour quality and quantity due to volatilisation of the volatile organic compounds present in the effluent. Remediation of these components was carried out by Fenton oxidation prior to evaporation. A high removal was observed under the applied conditions of Fe^{2+} = 25 mg/L, H_2O_2 = 50 mg/L, pH = 3 and a reaction time of 2 hours.

Second, a novel filtrative/adsorptive hybrid reactor termed Biological Membrane Assisted Carbon Filtration (BioMAC) was proposed for polishing of wastewater treatment plant effluent. It was observed that the reactor could be operated under stable flux conditions although regular hydraulic backwashing was necessary. Preferential attached growth of nitrifiers on the activated carbon particles allowed for a complete and very stable nitrification, with permeate total ammonium nitrogen and nitrite levels below 0.2 mg/L regardless of influent concentrations. Chemical Oxygen Demand of the permeate averaged 5.26 mg O_2/L. Elimination of volatile compounds was assessed using an electronic nose. The combined process resulted in complete odour removal, with permeate odour levels equalling the reference samples (tap water), even during periods of increased reactor load (shock load experiment). A 4.2 $\log_{10}$CFU and 3.7 $\log_{10}$CFU removal were observed for total coliforms and *E. coli*, respectively.

A third experimental part studied the wastes generated by RO filtration processes, *i.e.* the membrane concentrates. Indeed, the cost of disposal or treatment of membrane concentrates is rapidly becoming one of the main factors influencing the feasibility of any membrane application. As biological tests using the BioMAC reactor illustrated the very poor biodegradability of the RO brine, research efforts were focussed on

electrolysis as a novel treatment approach. Four different materials have been tested as anode for a number of brine samples in a one-compartment electrolytic cell in galvanostatic mode. It was found that PbO_2 and SnO_2 anodes initiated electrochemical precipitation through an increase of the pH. Boron-doped diamond and RuO_2 coated Ti anodes successfully oxidised the pollutants in the brine and a linear removal of total ammonia nitrogen (TAN) and chemical oxygen demand (COD) was observed during the first phase of oxidation. Oxidation was predominantly achieved through indirect hypochlorite bulk oxidation; the higher oxidation rate and extent for the boron-doped diamond anode was attributed to the higher selectivity and activity of the latter. Overall performance of the boron-doped diamond electrode was higher than for RuO_2 anodes: higher rates for TAN (17.9 vs. 13.5 mg/Ah) and COD (74.5 vs. 20.0 mg/Ah) removal as well as superior overall current efficiencies (35.2 vs. 14.5 %) were noted. Extensive colour removal was observed for both anodes (> 90 % decrease in absorbency at 455 nm).

Current and future developments in the field of advanced wastewater treatment and water reclamation indicate an increasing dominance of membrane technology. Filtrative separation is becoming increasingly cost-effective (*e.g.* development of novel membrane materials and larger membrane modules), while it seems to offer the assurance of efficiently retaining emerging pollutants, such as pharmaceuticals and endocrine disrupting chemicals. However, this emphasises the importance of membrane concentrate treatment, which will inevitably become a major point of focus. Therefore, the development of a reliable and cost-efficient concentrate abatement technology is of paramount importance to allow sustainable reuse of municipal wastewater. In this respect, the promising results obtained for the electro-oxidation of RO concentrates merit further research.

SAMENVATTING

Water is onze belangrijkste en meest waardevolle levenselement. Er zijn echter meer en meer aanwijzingen dat het een schaars goed wordt, ook in landen met een schijnbaar voldoende grote watervoorraad. Ondanks het gematigde zeeklimaat en meer dan 800 mm jaarlijkse neerslag heeft België slechts een beperkte hoeveelheid beschikbaar water, waarvan momenteel reeds meer dan 40 % benut wordt. Daarenboven hebben over-extractie van het grondwater en vervuiling van grond- en oppervlaktewaters geleid tot een dalende kwaliteit van het nog beschikbare water. Deze enorme menselijk impact op kwaliteit en kwantiteit van de waterreserves toont de noodzaak van een duurzaam waterbeleid overduidelijk aan: er dient gezorgd te worden voor een dynamische balans tussen vraag en aanbod. Vermits in veel regio's de bestaande watervoorraden maximaal benut worden, kan een stijgende vraag enkel beantwoord worden door alternatieve waterbronnen aan te spreken. In stedelijke agglomeraties en andere dichtbevolkte gebieden is het geproduceerde afvalwater een te overwegen "nieuwe" waterbron.

Hergebruik en recyclage van afvalwater geven aanleiding tot het ontstaan van korte cycli in de natuurlijke hydrologische cyclus, waardoor een bijna eindeloze "nieuwe" waterbron bekomen wordt. Eén van deze beschikbare bronnen is het effluent van rioolwaterzuiveringsinstallaties (RWZIs). In theorie kan water hergebruikt worden voor welke toepassing ook, enkel de vereiste behandeling verschilt. Er wordt onderscheid gemaakt tussen zeven types hergebruik: landbouwtoepassingen, stedelijke irrigatie, industriële toepassingen, grondwaterheraanvulling, milieutoepassingen en direct of indirect hergebruik voor drinkwater of andere doeleinden. RWZI effluent kan als dusdanig ingezet worden voor bepaalde toepassingen, zoals irrigatie in de landbouw, maar vereist meestal extra behandeling vooraleer het kan hergebruikt worden. Bij vele toepassingen is een gedeeltelijke of gehele verwijdering van de microbiologische, chemische en organoleptische componenten noodzakelijk om ongewenste effecten (zoals corrosie, verstopping of fouling van installaties) te vermijden, maar vooral ook om de volksgezondheid te beschermen. Daarom wordt in veel reglementeringen betreffende hergebruik de vereiste kwaliteit gekoppeld aan de beoogde toepassing.

Hergebruiktoepassingen zoals onrechtstreekse drinkwaterbereiding of stedelijke toepassingen vereisen meestal één of andere vorm van tertiaire behandeling, waarbij een enorme variatie aan technologieën beschikbaar is. Het doel van dit werk bestond erin geschikte technieken voor de verregaande zuivering van effluent te onderzoeken en te beoordelen. Hierbij werd de nadruk gelegd op membraanfiltratie, verdamping en elektrolyse.

In het eerste experimenteel deel werd verdamping onderzocht als een mogelijk alternatief voor membraanfiltratie. Verdamping van effluent geeft een evaporaat dat een gelijkaardige (of zelfs betere) kwaliteit heeft dan permeaat van een omgekeerde osmose installatie. Bij een pH lager dan 4 kon vervluchtiging van ammonium vermeden worden, doch dit gaf aanleiding tot de vervluchtiging van nitriet door de vorming van stikstofoxide. Daarenboven zorgde vervluchtiging van vluchtige organische componenten voor een toename van de geurhoeveelheid en een verandering in de geurkwaliteit van het evaporaat. Een Fenton voorbehandeling (Fe^{2+} = 25 mg/L, H_2O_2 = 50 mg/L; pH = 3; reactietijd = 2 uur) resulteerde in een volledige eliminatie van deze componenten, terwijl de lage pH ook het ammonium in het concentraat kon weerhouden.

In een tweede deel werd een hybride filtratie / adsorptie reactor, Biological Membrane Assisted Carbon filtration (BioMAC) getest. De flux doorheen de membranen kon stabiel gehouden worden, maar regelmatig terugspoelen met permeaat of lucht was nodig. De actieve koolpartikels gaven aanleiding tot een preferentiële groei van nitrificerende bacteriën en bijgevolg tot een stabiele en volledige nitrificatie. De permeaat TAN en nitrietconcentraties bleven lager dan 0.2 mg/L, onafhankelijk van de influentconcentraties. De chemische zuurstofvraag van het permeaat bedroeg gemiddeld 5.26 mg O_2/L. De verwijdering van vluchtige componenten werd bepaald door middel van een elektronische neus. Het geteste hybride proces gaf aanleiding tot een volledige verwijdering van geur, wat bleek uit de gelijkenis tussen de geurhoeveelheid in het permeaat en de referentiestalen, ook tijdens een experiment met piekbelasting. Verder werd een 4.2 $\log_{10}$ KVE en 3.7 $\log_{10}$ KVE verwijdering van totale coliformen en *E. coli* vastgesteld.

Een derde hoofdstuk bestudeerde het afval gegenereerd door membraanfiltratie processen, meer bepaald de membraanconcentraten. Immers, lozing of behandeling van deze concentraten is voor veel membraantoepassingen een doorslaggevende factor. Biologische tests op de BioMAC reactor gaven aan dat de concentraten zeer slecht biodegradeerbaar waren. Daarom werd elektrolyse als een alternatieve behandelingsstrategie naar voor geschoven. In een niet-gescheiden elektrolysecel werden vier verschillende elektrodematerialen getest voor verschillende membraanconcentraten (galvanostatische batch experimenten). PbO_2 en SnO_2 gaven aanleiding tot precipitatie door een toename van de pH. Boor gedopeerde diamant en RuO_2 elektrodes waren in staat de aanwezige polluenten (ammonium, chemische zuurstofvraag en kleur) te oxideren. Tijdens de eerste fase van de elektrochemische oxidatie werden vooral hypochloriet-ionen gevormd, hetgeen resulteerde in een lineaire daling van de concentratie van de polluenten. In een tweede fase van de oxidatie werd de generatie van vrije chloor waargenomen. In het algemeen was de diamant elektrode performanter dan de RuO_2: hogere verwijderingssnelheden voor ammoniumstikstof (17.9 tegenover 13.5 mg/Ah) en chemische zuurstofvraag (74.5 tegenover 20.0 mg/Ah) en betere stroomefficiëntie (35.2 tegenover 14.5 %) werden gemeten. Elektrolyse resulteerde in beide gevallen in een volledige kleurverwijdering (> 90 % daling in absorbantie bij 455 nm).

De huidige en voorspelde ontwikkelingen in membraantechnologie zullen aanleiding geven tot een toenemende dominantie van deze techniek. Scheiding door membraanfiltratie is in toenemende mate kosten-efficiënt (o.a. door de ontwikkeling van nieuwe membraanmaterialen en grotere membraanmodules), terwijl blijkbaar ook nieuw types polluenten, zoals medicijnresiduen en pseudo-eustrogenen zonder problemen verwijderd worden. Anderzijds zal hierdoor een nieuw afvalproduct, meer bepaald het geproduceerde membraanconcentraat, steeds belangrijker worden. Een duurzame ontwikkeling van membraanfiltratie als een proces voor afvalwaterzuivering en hergebruik zal dan ook enkel mogelijk zijn indien het gekoppeld wordt aan een efficiënte en betrouwbare techniek voor concentraatbehandeling.

BIBLIOGRAPHY

Abdel-Jawad M., Ebrahim S., Al-Tabtabaei M. and Al-Shammari S. (1999) Advanced technologies for municipal wastewater purification: technical and economic assessment. *Desalination*, **124** (1-3), 251-261.

Abdessemed D., Nezzal G. and Ben Aim R. (2000) Coagulation-adsorption-ultrafiltration for wastewater treatment and reuse. *Desalination*, **131** (1-3), 307-314.

Ahmed M., Shayya W. H., Hoey D. and Al-Handaly J. (2001) Brine disposal from reverse osmosis desalination plants in Oman and the United Arab Emirates. *Desalination*, **133** (2), 135-147.

Ahmed M., Shayya W. H., Hoey D., Mahendran A., Morris R. and Al-Handaly J. (2000) Use of evaporation ponds for brine disposal in desalination plants. *Desalination*, **130** (2), 155-168.

Ahn K. H. and Song K. G. (1999) Treatment of domestic wastewater using microfiltration for reuse of wastewater. *Desalination*, **126** (1-3), 7-14.

Ahn K. H. and Song K. G. (2000) Application of microfiltration with a novel fouling control method for reuse of wastewater from a large-scale resort complex. *Desalination*, **129** (3), 207-216.

AHPA (1992) *Standard methods for the examination of water and wastewater*. Greenberg A. E., Clesceri L. S. and Eaton A. D. (eds.), 18 th edition, American Public Health Association, Washington, D.C.

Alpha M.O.S. (1995) *FOX 2000 to 4000 Electronic Nose User Manual*. Alpha M.O.S., Toulouse.

Al-Shammiri M. and Safar M. (1999) Multi-effect distillation plants: state of the art. *Desalination*, **126** (1-3), 45-59.

Amahmid O., Asmama S. and Bouhoum K. (1999) The effect of waste water reuse in irrigation on the contamination level of food crops by *Giardia* cysts and *Ascaris* eggs. *International Journal of Food Microbiology*, **49** (1-2), 19-26.

Anda M., Mathew K. and Ho G. (2001) Evapotranspiration for domestic wastewater reuse in remote indigenous communities of Australia. *Water Science and Technology*, **44** (6), 1-10.

Anderson J. (2001) Prospects for international guidelines for water recycling. *Water 21*, **3** (8), 16-21.

Anderson J., Adin A., Crook J., Davis C., Hultquist R., Jimenez-Cisneros B., Kennedy W., Sheikh B. and van der Merwe B. (2001) Climbing the ladder: a step by step approach to international guidelines for water recycling. *Water Science and Technology*, **43** (10),1-9.

Andreozzi R., Marotta R., Pinto G. and Pollio A. (2001) Carbamazepine in water: persistence in the environment, toxicity towards algae and ozonation treatment. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Anonymous (1999) Grootschalig onderzoek naar hergebruik effluent. *H_2O*, **32** (1), 4-5.

Anonymous (2000) Hergebruik zal waterschaarste in Vlaanderen moeten oplossen. *Aqua, driemaandelijks informatieblad van Aquafin*, (3), 2-3.

Anpilov A. M., Barkhudarov E. M., Bark Y. B., Zadiraka Y. V., Christofi M., Kozlov Y. N., Kossyi I. A., Kop'ev V. A., Silakov V. P., Taktakishvili M. I. and Temchin S. M. (2001) Electric discharge in water as a source of UV radiation, ozone and hydrogen peroxide. *Journal of Physics D-Applied Physics*, **34** (6), 993-999.

Asano T. (1994) Reusing urban wastewater - an alternative and reliable water resource. *Water International*, **19** (1), 36-42.

Asano T. and Levine A. D. (1995) Wastewater reuse: a valuable link in water resources management. *Water Quality International* (4), 20-24.

Asano T. and Levine A. D. (1998) Wastewater reclamation, recycling and reuse: an introduction. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 1-56.

Atlas R. M. (1993) *Handbook of microbiological media*. Parks L. C. (ed.), CRC Press, Boca Raton, Florida.

Bahman S., Cort R., Cooper R. C. and Jaques R. S. (1998) Tertiary-treated reclaimed water for irrigation of raw-eaten vegetables. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 779-825.

Bahri A. (1998) Wastewater reclamation and reuse in Tunesia. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 877-916.

Baker R. W. (1998) Membrane technology. *In: Kirk-Othmer Encyclopaedia of Chemical Technology*. 4 th edition, John Wiley & Sons, New York, 135-193.

Balanosky E., Fernandez J., Kiwi J. and Lopez A. (1999) Degradation of membrane concentrates of the textile industry by Fenton like reactions in iron-free solutions at biocompatible pH values (pH approximate to 7-8). *Water Science and Technology*, **40** (4-5), 417-424.

Bannoud A. H., Persin F. and Rumeau M. (1993) A study of the perfection of an electrochemical reactor for softening water. *Water Research*, **27** (8), 1385-1391.

Baumgarten G. and Seyfried C. F. (1996) Experiences and new developments in biological pre-treatment and physical post-treatment of landfill leachate. *Water Science and Technology*, **34** (7-8), 445-453.

Beaufays J.-P. and Dubois P. (1999) Gestion durable des resources en eau. *Annales Medicinae Militaris Belgicae*, **13** (1-2), 159-167.

Benedek A. (2001) The evolution of membrane technology and its potential for alleviating global water problems. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Bilstad T. (1997) Membrane operations. *Water Science and Technology*, **36** (2-3), 17-24.

Birchler D. R., Milke M. W., Marks A. L. and Luthy R. G. (1994) Landfill leachate treatment by evaporation. *Journal of Environmental Engineering*, **120** (5), 1109-1131.

Birkett J. (2000) Water reuse: finding water for a growing population. *In: World of Water 2000*. Penn Well, Tulsa, Oklahoma, 136-140.

Blackson D. E. and Moreland J. L. (1998) Wastewater reclamation ad reuse for cooling towers at the Palo Verde nuclear generating station. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 1143-1161.

Blumenthal U. J., Duncan Mara D., Peasey A., Ruiz-Palacios R. and Stott R. (2000) Guidelines for the microbiological quality of treated wastewater used in agriculture: recommendations for revising WHO guidelines. *Bulletin of the World Health Organisation*, **78** (9), 1104-1116.

Bonfatti F., De Battisti A., Ferro S., Lodi G. and Osti S. (2000a) Anodic mineralisation of organic substrates in chloride-containing aqueous media. *Electrochimica Acta*, **46** (2-3), 305-314.

Bonfatti F., Ferro S., Lavezzo F., Malacarne M., Lodi G. and De Battisti A. (1999) Electrochemical incineration of glucose as a model organic substrate I. Role of the electrode material. *Journal of the Electrochemical Society*, **146** (6), 2175-2179.

Bonfatti F., Ferro S., Lavezzo F., Malacarne M., Lodi G. and De Battisti A. (2000b) Electrochemical incineration of glucose as a model organic substrate - II. Role of active chlorine mediation. *Journal of the Electrochemical Society*, **147** (2), 592-596.

Bonnélye V., Colas F., Levasseur W., Abidine N., Dimitriou M. A. and Roux J. P. (2001) Adaptation of the ultrafiltration process to huge plant scale. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Bontoux L. (1998) The regulatory status of wastewater reuse in the European Union. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 1463-1475.

Borsani R., Fazio M. and Ferrando B. (1996) Industrial water production by utilisation of a reverse osmosis and an evaporation plant. *Desalination*, **108** (1-3), 231-245.

Braunstein J. L., Loge F. J., Tchobanoglous G. and Darby J. L. (1996) Ultraviolet disinfection of filtered activated sludge effluent for reuse applications. *Water Environment Research*, **68** (2), 152-161.

Breschi D. (2000) Design changes improve MSF efficiency. *Water and Wastewater International*, **14** (5), 25-27.

Bruchet A., Prompsy C., Filippi G. and Souali A. (2001) A broad spectrum analytical scheme for the screening of endocrine disruptors (ED's), pharmaceuticals and personal care products in wastewaters and natural waters. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Buros O. K. (2000) *The ABCs of desalting*. International Desalination Organisation, Topsfield, Massachusetts, USA.

Buyuksonmez F., Hess T. F., Crawford R. L. and Watts R. J. (1998) Toxic effects of modified Fenton reactions on *Xanthobacter flavus* FB71. *Applied and Environmental Microbiology*, **64** (10), 3759-3764.

Campos C., Marinas B. J., Snoeyink V. L., Baudin I. and Laine J. M. (2000a) PAC-membrane filtration process. I: Model development. *Journal of Environmental Engineering*, **126** (2), 97-103.

Campos C., Marinas B. J., Snoeyink V. L., Baudin I. and Laine J. M. (2000b) PAC-membrane filtration process. II: Model application. *Journal of Environmental Engineering*, **126** (2), 104-111.

Campos C., Schimmoller L., Marinas B. J., Snoeyink V. L., Baudin I. and Laine J.-M. (2000c) Adding PAC to remove DOC. *Journal of the American Water Works Association*, **92** (8), 69-83.

Casey E., Glennon B. and Hamer G. (1999) Oxygen mass transfer characteristics in a membrane-aerated biofilm reactor. *Biotechnology and Bioengineering*, **62** (2), 183-192.

Chaibi M. T. (2000) An overview of solar desalination for domestic and agriculture water needs in remote arid areas. *Desalination*, **127** (2), 119-133.

Chang A. C., Page A. L., Asano T. and Hespanhol I. (1998) Evaluating methods of establishing human health-related chemical guidelines for cropland application of municipal wastewater. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 705-756.

Chen W. and Horan J. (1998) The treatment of a high strength pulp and paper mill effluent for wastewater reuse - IV) A pilot study into the production of high quality recycle water using tertiary and deep treatment. *Environmental Technology*, **19** (9), 861-871.

Chiang L. C., Chang J. E. and Wen T. C. (1995) Indirect oxidation effect in electrochemical oxidation treatment of landfill leachate. *Water Research*, **29** (2), 671-678.

Choi J. and Valentine R. L. (2002) Formation of N-nitrosodimethylamine (NDMA) from reaction of monochloramine: a new disinfection by-product. *Water Research*, **36** (4), 817-824.

Ciardelli G., Corsi L. and Marcucci M. (2001) Membrane separation for wastewater reuse in the textile industry. *Resources, Conservation and Recycling*, **31** (2), 189-197.

Cicek N., Franco J. P., Suidan M. T. and Urbain V. (1998) Using a membrane bioreactor to reclaim wastewater. *Journal of the American Water Works Association*, **90** (11), 105-113.

Comninellis C. (1994) Electrocatalysis in the electrochemical conversion/combustion of organic pollutants for wastewater treatment. *Electrochimica Acta*, **39** (11-12), 1857-1862.

Comninellis C. and Nerini A. (1995) Anodic oxidation of phenol in the presence of NaCl for wastewater treatment. *Journal of Applied Electrochemistry*, **25** (1), 23-28.

Comninellis C. and Pulgarin C. (1991) Anodic oxidation of phenol for wastewater treatment. *Journal of Applied Electrochemistry*, **21** (8), 703-708.

Comninellis C. and Pulgarin C. (1993) Electrochemical oxidation of phenol for wastewater treatment using SnO_2 anodes. *Journal of Applied Electrochemistry*, **23** (2), 108-112.

Cooper R. C. and Olivieri A. W. (1998) Infectious disease concerns in wastewater reuse. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 489-520.

Côté P., Buisson H., Pound C. and Arakaki G. (1997) Immersed membrane activated sludge for the reuse of municipal wastewater. *Desalination*, **113** (2-3), 189-196.

Côté P., Cadera J., Adams N. and Mende U. (2001) A new immersed membrane for water filtration. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Crook J. (1998) Water reclamation and reuse criteria. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 627-703.

Crook J., MacDonald J. A. and Trussell R. R. (1999) Potable use of reclaimed water. *Journal of the American Water Works Association*, **91** (8), 40-49.

De Ceukelaire K., Walraevens K., Van Brum P. and De Breuck W. (1992) Evolution of equilibrium heights in the Landeniaan and the Sokkel from 1900 to 1986 (West Flanders and the neighbouring part of East-Flanders). *Water*, **65** (4), 113-117.

Dellamonica M., Agostiano A. and Ceglie A. (1980) An electrochemical sewage treatment process. *Journal of Applied Electrochemistry*, **10** (4), 527-533.

Dewettinck T. (2002) *Monitoring and controlling the quality of reclaimed domestic wastewater*. Ph.D. thesis in preparation, Ghent University.

Dewettinck T., Van Hege K., Tanghe T. and Verstraete W. (1999) Doorgedreven zuivering van afvalwater voor hergebruik in industriële processen. *Annales Medicinae Militaris Belgicae*, **13** (1-2), 43-48.

Dewettinck T., Van Hege K. and Verstraete W. (2001a) Development of a rapid pH-based biosensor to monitor and control the hygienic quality of reclaimed domestic wastewater. *Applied Microbiology and Biotechnology*, **56** (5-6), 809-815.

Dewettinck T., Van Hege K. and Verstraete W. (2001b) The electronic nose as a rapid sensor for volatile compounds in treated domestic wastewater. *Water Research*, **35** (10), 2475-2483.

Downing J. B., Bracco A., Green F. B., Ku A. Y., Lundquist T. J. and Zubieta I. X. (2002) Low cost reclamation using the advanced integrated wastewater pond systems technology and reverse osmosis. *Water Science and Technology*, **45** (1), 117-125.

Drewes J. E., Heberer T. and Reddersen K. (2001) Fate of pharmaceuticals during indirect potable reuse. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Drewes J. E., Heberer T. and Reddersen K. (2002) Removal of pharmaceuticals during conventional wastewater treatment, advanced membrane treatment and soil aquifer treatment. *Water Resources submitted*.

Duin O., Wessels P., van der Roest H., Uijterlinde C. and Schoonewille H. (2000) Direct nanofiltration or ultrafiltration of WWTP effluent? *Desalination*, **132** (1-3), 65-72.

EEA (1995) *Europe's environment. The Dobris assessment*. European Environment Agency, Copenhagen.

EEA (1997) *Water resources problems in Southern Europe - An overview report.* Topic Report 15/1997, European Environmental Agency, Copenhagen.

EEA (1998) *Europe's Environment. The second assessment*. European Environmental Agency, Copenhagen.

EEA (1999a) *Environment in the European Union at the turn of the century*. Environmental assessment report No 2, European Environmental Agency, Copenhagen.

EEA (1999b) *Sustainable water use in Europe. Part 1: sectoral use of water*. Environmental assessment report No 1, European Environmental Agency, Copenhagen.

EEA (2000) *Sustainable use of Europe's water ? State, prospect and issues*. Environmental assessment series No 7, European Environment Agency, Copenhagen.

EEA/UNEP (1997) *Water stress in Europe - can the challenge be met?* European Environmental Agency / United Nations Environment Program (EEA / UNEP), Copenhagen.

Ericsson B. and Hallmans B. (1996) Treatment of saline wastewater for zero discharge at the Debiensko coal mines in Poland. *Desalination*, **105** (1-2), 115-123.

ETC/IW (1998) *Groundwater quality and quantity in Europe*. EEA monograph P026/97/1, European Topic Centre on Inland Waters, Copenhagen.

European Commission (1991) *Council directive concerning urban wastewater treatment*. 91/271/EEC, European Commission.

European Commission (2000) *Framework for Community action in the flied of water policy*. 2000/60/EC, European Parliament and the Council.

Facey R. M. and Smith D. W. (2001) Freeze-thaw treatment of membrane concentrates derived from Kraft pulp mill operations. *Journal of Cold Regions Engineering*, **15** (2), 69-90.

Fane A. G. (1996) Membranes for water production and wastewater reuse. *Desalination*, **106** (1-3), 1-9.

Fane A. G., Beatson P. and Li H. (2000) Membrane fouling and its control in environmental applications. *Water Science and Technology*, **41** (10-11), 303-308.

Fenton H. J. H. (1894) Oxidation of tartaric acid in presence of iron. *Journal of the Chemical Society*, **65**, 899-910.

Ferro S., De Battisti A., Duo I., Comninellis C., Haenni W. and Perret A. (2000) Chlorine evolution at highly boron-doped diamond electrodes. *Journal of the Electrochemical Society*, **147** (7), 2614-2619.

Filteau G., Whitley C. and Watson I. C. (1995) Water reclamation fuels economic growth in Harlingen, Texas. Reclamation of municipal wastewater for industrial process use. *Desalination*, **103** (1-2), 31-37.

Flemming H. C., Schaule G., Griebe T., Schmitt J. and Tamachkiarowa A. (1997) Biofouling - the Achilles heel of membrane processes. *Desalination*, **113** (2-3), 215-225.

Ford D. L. (1981) Wastewater characteristics and treatment. *In: Activated carbon adsorption for wastewater treatment*. Perrich J. R. (ed.), CRC Press, Boca Raton, Florida, 1-27.

Foti G., Gandini D., Comninellis C., Perret A. and Haenni W. (1999) Oxidation of organics by intermediates of water discharge on IrO_2 and synthetic diamond anodes. *Electrochemical and Solid-State Letters*, **2** (5), 228-230.

Gagné F. and Blaise C. (1998) Estrogenic properties of municipal and industrial wastewaters evaluated with a rapid and sensitive chemoluminescent in situ hybridization assay (CISH) in rainbow trout hepatocytes. *Aquatic Toxicology*, **44** (1-2), 83-91.

Gander M., Jefferson B. and Judd S. (2000a) Aerobic MBRs for domestic wastewater treatment: a review with cost considerations. *Separation and Purification Technology*, **18** (2), 119-130.

Gander M. A., Jefferson B. and Judd S. J. (2000b) Membrane bioreactors for use in small wastewater treatment plants: membrane materials and effluent quality. *Water Science and Technology*, **41** (1), 205-211.

Gandini D., Mahe E., Michaud P. A., Haenni W., Perret A. and Comninellis C. (2000) Oxidation of carboxylic acids at boron-doped diamond electrodes for wastewater treatment. *Journal of Applied Electrochemistry*, **30** (12), 1345-1350.

Gernaey K., Verschuere L., Luyten L. and Verstraete W. (1997) Fast and sensitive acute toxicity detection with an enrichment nitrifying culture. *Water Environment Research*, **69** (5), 1163-1169.

Gherardini L., Michaud P. A., Panizza M., Comninellis C. and Vatistas N. (2001) Electrochemical oxidation of 4-chlorophenol for wastewater treatment - Definition of normalised current efficiency (phi). *Journal of the Electrochemical Society*, **148** (6), D78-D82.

Ghyoot W. and Verstraete W. (2000) Reduced sludge production in a two-stage membrane-assisted bioreactor. *Water Research*, **34** (1), 205-215.

Gleick P. H. (2000) Water reclamation and reuse: waste not, want not. *In: The World's water 2000-2001: the biennial report on freshwater resources*. Island press, Washington, D.C.

Gnirss R. (2000) Microfiltration of municipal wastewater in Berlin : Experience of operation and cost evaluation. *In: 4th Biennial Congress of the African Division of the International Association of Hydraulic Research*, 7 - 9 June 2000. IAHR, Windhoek, Namibia.

Gostelow P., Parsons S. A. and Stuetz R. M. (2001) Odour measurements for sewage treatment works. *Water Research*, **35** (3), 579-597.

Goto M. and Kuribayashi S. (2001) Behaviour of endocrine disrupting chemicals in wastewater treatment plants. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Grimm J., Bessarabov D. and Sanderson R. (1998) Review of electro-assisted methods for water purification. *Desalination*, **115** (3), 285-294.

Haarhoff J. and van der Merwe B. (1996) Twenty-five years of wastewater reclamation in Windhoek, Namibia. *Water Science and Technology*, **33** (10-11), 25-35.

Hartling E. C. and Nellor M. H. (1998) Water recycling in Los Angeles County. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 917-940.

Heberer T., Reddersen K. and Mechlinski A. (2001) From municipal sewage to drinking water: fate and removal of pharmaceutical residues in the aquatic environment in urban areas. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Howell J. A. (1995) Subcritical flux operation of microfiltration. *Journal of Membrane Science*, **107** (1-2), 165-171.

Huang C. H. and Sedlak D. L. (2001) Analysis of estrogenic hormones in municipal wastewater effluent and surface water using enzyme-linked immunosorbent assay and gas chromatography/tandem mass spectrometry. *Environmental Toxicology and Chemistry*, **20** (1), 133-139.

Huang Y. H., Chou S. S., Perng M. G., Huang G. H. and Cheng S. S. (1999) Case study on the bio-effluent of petrochemical wastewater by electro-Fenton method. *Water Science and Technology*, **39** (10-11), 145-149.

Husain H. and Côté P. (1999) Membrane bioreactors for municipal wastewater treatment. *Water Quality International* (2), 19-22.

Hussain G. and Al-Saati A. J. (1999) Wastewater quality and its reuse in agriculture in Saudi Arabia. *Desalination*, **123** (2-3), 241-251.

Huston P. L. and Pignatello J. J. (1999) Degradation of selected pesticide active ingredients and commercial formulations in water by the photo-assisted Fenton reaction. *Water Research*, **33** (5), 1238-1246.

Iniesta J., González-García J., Expósito E., Montiel V. and Aldaz A. (2001a) Influence of chloride ion on electrochemical degradation of phenol in alkaline medium using bismuth doped and pure PbO_2 anodes. *Water Research*, **35** (14), 3291-3300.

Iniesta J., Michaud P. A., Panizza M. and Comninellis C. (2001b) Electrochemical oxidation of 3-methylpyridine at a boron-doped diamond electrode: application to electro-organic synthesis and wastewater treatment. *Electrochemistry Communications*, **3** (7), 346-351.

IPCC (1996) *Climate change 1995: 'The science of climate change', contribution of working group I to the second assessment report of the Interngovernmental Panel on Climate Change.* Cambridge University Press, Cambridge.

Irvine E., Bavister G., Welch D. and Rachwal T. (2001) An update on experience of nanofiltration for colour removal in remote scottish communities. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Janssens I., Tanghe T. and Verstraete W. (1997) Micropollutants: a bottleneck in sustainable wastewater treatment. *Water Science and Technology*, **35** (10), 13-26.

Jefferson B., Laine A. L., Judd S. L. and Stephenson T. (2000) Membrane bioreactors and their role in wastewater reuse. *Water Science and Technology*, **41** (1), 197-204.

Johnson W. D. and Parnell J. R. (1998) Wastewater reclamation and reuse in the city of St-Petersburg, Florida. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 1037-1104.

Johnson W. T., Phelps R. W., Beatson P. J. and Fane A. G. (1997) Wastewater reuse using membranes. *In: IDA World Congress on Desalination and Water Reuse*, 6-9 October 1997. International Desalination Association, Madrid, Spain, 611-628.

Jolis D., Campana R., Hirano R., Pitt P. and Marinas B. (1995) Desalination of municipal wastewater for horticultural reuse: process description and evaluation. *Desalination*, **103** (1-2), 1-10.

Jolis D., Hirano R. and Pitt P. (1999) Tertiary treatment using microfiltration and UV disinfection for water reclamation. *Water Environment Research*, **71** (2), 224-231.

Judd S. (1999) Industry's reuse challenge. *Water 21*, **1** (5), 48-49.

Juttner K., Galla U. and Schmieder H. (2000) Electrochemical approaches to environmental problems in the process industry. *Electrochimica Acta*, **45** (15-16), 2575-2594.

Kanarek A., Aharoni A. and Michail M. (1993) Municipal wastewater reuse via soil aquifer treatment for non-potable purposes. *Water Science and Technology*, **27** (7), 53-61.

Kanarek A. and Michail M. (1996) Groundwater recharge with municipal effluent : Dan region reclamation project, Israel. *Water Science and Technology*, **34** (11), 227-233.

Khan S. J. and Ongerth J. E. (2001) Estimation of pharmaceutical residues in primary and secondary sewage sludge based on quantities of use and fugacity modelling. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Khow J. H., Smith A. J., Rachwell A., Donn A. and Meadowcroft C. V. (2000) Water reuse for the next millennium - membrane treatment at the millennium dome. *In: Membrane technology in water and wastewater treatment*. Hillis P. (ed.), Royal society of chemistry, London, 175-183.

Kim J., Gonzalez-Martin A., Salinas C. and Rutherford L. A. (2001) Electrochemical removal of ammonium ions from a bioreactor effluent. *Life Support and Biosphere Science*, **8** (1), 23-31.

Kim J. S., Lee C. H. and Chun H. D. (1998) Comparison of ultrafiltration characteristics between activated sludge and BAC sludge. *Water Research*, **32** (11), 3443-3451.

KMI (2002) *Klimatologische normalen voor Ukkel 1961-1990*. Koninklijk Meteorologisch Instituut, http://www.meteo.be, Ukkel, Belgium.

Kochany J. and Lugowski A. (1998) Application of Fenton's reagent and activated carbon for removal of nitrification inhibitors. *Environmental Technology*, **19** (4), 425-429.

Koistinen P. R. (1999) *New evaporation treatment of industrial effluents and landfill leachates using technology with polymeric heat transfer surfaces*. Hadwaco Inc., Helsinki, Finland.

Kraft A., Blaschke M., Kreysig D., Sandt B., Schroder F. and Rennau J. (1999a) Electrochemical water disinfection - Part II: Hypochlorite production from potable water, chlorine consumption and the problem of calcareous deposits. *Journal of Applied Electrochemistry*, **29** (8), 895-902.

Kraft A., Stadelmann M., Blaschke M., Kreysig D., Sandt B., Schroder F. and Rennau J. (1999b) Electrochemical water disinfection - Part I: Hypochlorite production from very dilute chloride solutions. *Journal of Applied Electrochemistry*, **29** (7), 861-868.

Kuo J. F., Stahl J. F., Chen C. L. and Bohlier P. V. (1998) Dual role of activated carbon process for water reuse. *Water Environment Research*, **70** (2), 161-170.

Laine J., Bruchet A., Jacangelo J. G., Glucina K. and Baudin I. (2000) Assessment of membrane processes for taste and odour removal. *In: Proceedings of the 1st world water congress of the International Water Association*, 3 - 7 July 2000. IWA, Paris.

Larsen T. A. and Gujer W. (1997) The concept of sustainable urban water management. *Water Science and Technology*, **35** (9), 3-10.

Lazarova V., Levine B., Sack J., Cirelli G., Jeffrey P., Muntau H., Brissaud F. and Salgot M. (2000) Role of water reuse in enhancement of integrated water management in Europe and Mediterranean countries. *In: 3rd International Symposium on Wastewater, Reclamation, Recycling and Reuse (WRRR)*, 3-7 July 2000. International Water Association (IWA), Paris.

Lebeau T., Lelièvre C., Buisson H., Cleret D., Van de Venter L. W. and Côté P. (1998) Immersed membrane filtration for the production of drinking water: combination with PAC for NOM and SOCs removal. *Desalination*, **117** (1-3), 219-231.

Li X. Z. and Zhao Y. G. (1999) Advanced treatment of dyeing wastewater for reuse. *Water Science and Technology*, **39** (10-11), 249-255.

Liessens J., Beeckman M., Janssens I. and Verstraete W. (1996) Wastewater treatment: recent developments particularly in relation to reuse. *In: Proceedings of the International Conference on Environmental Pollution*, 15-19 April 1996. Nath B., Lang I., Meszaros E., Robinson J. P. and Hens L. (eds.), European Centre for Pollution Research, Budapest, 24-35.

Lin S. H., Lin C. M. and Leu H. G. (1999) Operating characteristics and kinetic studies of surfactant wastewater treatment by Fenton oxidation. *Water Research*, **33** (7), 1735-1741.

Lin S. H. and Lin C. S. (1998) Reclamation of wastewater effluent from a chemical fiber plant. *Desalination*, **120** (3), 185-195.

Lin S. H., Shyu C. T. and Sun M. C. (1998) Saline wastewater treatment by electrochemical method. *Water Research*, **32** (4), 1059-1066.

Lin S. H. and Wu C. L. (1996) Electrochemical removal of nitrite and ammonia from aquaculture. *Water Research*, **30** (3), 715-721.

Livingston A. G., Arcangeli J. P., Boam A. T., Zhang S. F., Marangon M. and dos Santos L. M. F. (1998) Extractive membrane bioreactors for detoxification of chemical industry wastes: process development. *Journal of Membrane Science*, **151** (1), 29-44.

Madaeni S. S. (1999) The application of membrane technology for water disinfection. *Water Research*, **33** (2), 301-308.

Madireddi K., Babcock R. W. J., Levine B., Huo T. L., Khan E., Ye Q. F., Neethling J. B., Suffet I. H. and Stenstrom M. K. (1997) Wastewater reclamation at Lake Arrowhead, California: an overview. *Water Environment Research*, **69** (3), 350-362.

Madsen R. F. (1987) Membrane technology as a tool to prevent dangers to human health by water reuse. *Desalination*, **67**, 381-393.

Malik M. A., Ghaffar A. and Malik S. A. (2001) Water purification by electrical discharges. *Plasma Sources Science & Technology*, **10** (1), 82-91.

Marecos do Monte M. H. F. (1998) Agricultural irrigation with treated wastewater in Portugal. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 827-875.

Maricou H., Pereira D., Verschuere L., Philips S. and Verstraete W. (1998) Measurements of some volatile compounds by means of the electronic nose. *Water, Air and Soil Pollution*, **107** (1-4), 423-442.

Marincic L. and Leitz F. B. (1978) Electro-oxidation of ammonia in waste water. *Journal of Applied Electrochemistry*, **8**, 333-345.

Matsui Y., Colas F. and Yuasa A. (2001a) Removal of a synthetic organic chemical by PAC-UF systems. II: Model application. *Water Research*, **35** (2), 464-470.

Matsui Y., Yuasa A. and Ariga K. (2001b) Removal of a synthetic organic chemical by PAC-UF systems. I: Theory and modelling. *Water Research*, **35** (2), 455-463.

Matsunaga T., Okochi M., Takahashi M., Nakayama T., Wake H. and Nakamura N. (2000) TiN electrode reactor for disinfection of drinking water. *Water Research*, **34** (12), 3117-3122.

McEwen B. (1998) Indirect potable reuse of reclaimed water. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 1211-1267.

McKeough P. and Fagernas L. (1999) Further evaporation and final treatment of process water concentrates. *Water Science and Technology*, **40** (11-12), 25-32.

McMann B. (2002) Nuclear power's resource prospects. *Water 21*, **4** (4), 17-18.

Metcalf & Eddy (1991) *Wastewater engineering: treatment, disposal, and reuse*. McGraw-Hill Inc., New York.

Michaud P. A., Mahe E., Haenni W., Perret A. and Comninellis C. (2000) Preparation of peroxosulphuric acid using boron-doped diamond electrodes. *Electrochemical and Solid-State Letters*, **3** (2), 77-79.

Mills W. R., Bradford S. M., Rigby M. and Wehner M. P. (1998) Groundwater recharge at the Orange County Water District. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 1105-1142.

Miserez K., Philips S. and Verstraete W. (1999) New biology for advanced wastewater treatment. *Water Science and Technology*, **40** (4-5), 137-144.

Mitch W. A. and Sedlak D. L. (2002) Formation of N-nitrosodimethylamine (NDMA) from dimethylamine during chlorination. *Environmental Science and Technology*, **36** (4), 588-595.

Mobius C. H. and Cordes-Tolle M. (1994) Advanced treatment of paper mill wastewaters. *Water Science and Technology*, **29** (5-6), 273-282.

Morita C., Sano K., Morimatsu S., Kiura H., Goto T., Kohno T., Hong W., Miyoshi H., Iwasawa A., Nakamura Y., Tagawa M., Yokosuka O., Saisho H., Maeda T. and Katsuoka Y. (2000) Disinfection potential of electrolysed solutions containing sodium chloride at low concentrations. *Journal of Virological Methods*, **85** (1-2), 163-174.

Mujeriego R. and Asano T. (1999) The role of advanced treatment in wastewater reclamation and reuse. *Water Science and Technology*, **40** (4-5), 1-9.

Najm I. and Trussell R. R. (2001) NDMA formation in water and wastewater. *Journal of the American Water Works Association*, **93** (2), 92-99.

Nakazato T. and Kawamura Y. (1997) Improving the water environment with reclaimed wastewater. *Water Quality International* (6), 14-16.

Nghiem L. D., Schäfer A. I. and Waite T. D. (2001) Adsorption of estrone on NF and RO membranes in water and wastewater treatment. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Notsu H., Yagi I., Tatsuma, T., Tryk D.A. and Fujishima A. (2000) Surface carbonyl groups on oxidised diamond electrodes. *Journal of Electroanalytical Chemistry*, **492** (1), 31-37.

Odendaal P. E., van der Westhuizen J. L. J. and Grobler G. J. (1998) Wastewater reuse in South Africa. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 1163-1192.

Ogoshi M., Suzuki Y. and Asano T. (2001) Water reuse in Japan. *Water Science and Technology*, **43** (10), 17-25.

Okochi M., Lim T.-K., Nakamura N. and Matsunaga T. (1997) Electrochemical disinfection of drinking water using an activated-carbon-fiber reactor capable of monitoring its microbial fouling. *Applied Microbiology and Biotechnology*, **47** (1), 18-22.

Okun D. A. (2000) Water reclamation and unrestricted nonpotable reuse: a new tool in urban water management. *Annual Reviews of Public Health*, **21**, 223-245.

Okun D. A. (2001) Water reuse introduces the need to integrate both water supply and wastewater management at local and regulatory levels. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Oron G. (1996) Soil as a complementary treatment component for simultaneous wastewater disposal and reuse. *Water Science and Technology*, **34** (11), 243-252.

Oron G. (1998) Water resources management and wastewater reuse for agriculture in Israel. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 757-777.

Panizza M., Bocca C. and Cerisola G. (2000a) Electrochemical treatment of wastewater containing polyaromatic organic pollutants. *Water Research*, **34** (9), 2601-2605.

Panizza M., Duo I., Michaud P. A., Cerisola G. and Comninellis C. (2000b) Electrochemical generation of silver(II) at boron-doped diamond electrodes. *Electrochemical and Solid-State Letters*, **3** (12), 550-551.

Panizza M., Michaud P. A., Cerisola G. and Comninellis C. (2001a) Anodic oxidation of 2-naphthol at boron-doped diamond electrodes. *Journal of Electroanalytical Chemistry*, **507** (1-2), 206-214.

Panizza M., Michaud P. A., Cerisola G. and Comninellis C. (2001b) Electrochemical treatment of wastewaters containing organic pollutants on boron-doped diamond electrodes: prediction of specific energy consumption and required electrode area. *Electrochemistry Communications*, **3** (7), 336-339.

Park T. J., Lee K. H., Jung E. J. and Kim C. W. (1999) Removal of refractory organics and colour in pigment wastewater with Fenton oxidation. *Water Science and Technology*, **39** (10-11), 189-192.

Parmentier G., Thoeye C., De Gueldre G. and Norga K. (2000) Membraantechnologie en hergebruik van gezuiverd rioolwater in Vlaanderen: stand van zaken. *EcoTips* (6), 39-43.

Patermarakis G. and Fountoukidis E. (1990) Disinfection of water by electrochemical treatment. *Water Research*, **24** (12), 1491-1496.

Paxeus N. (1996) Organic pollutants in the effluents of large wastewater treatment plants in Sweden. *Water Research*, **30** (5), 1115-1122.

Pilkington N. H. (1995) Disinfection: an overview. *In: Modern techniques in water and wastewater treatment*. Kolarik L. O. and Priestley A. J. (eds.), CSIRO Australia, 75-79.

Pinkston K. E. and Sedlak D. L. (2001) The fate of pharmaceutically-active compounds in water recycling systems. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Postma H. J. W. (1982) Verwijdering en terugwinning van ammoniumstikstof uit afvalwater door toepassing van het lucht-strip proces. *H_2O*, **15** (16), 413-419.

Potter F. J. and Roth J. A. (1993) Oxidation of chlorinated phenols using Fenton reagent. *Hazardous Waste & Hazardous Materials*, **10** (2), 151-170.

Rajeshwar K., Ibanez J. G. and Swain G. M. (1994) Electrochemistry and the environment. *Journal of Applied Electrochemistry*, **24** (11), 1077-1091.

Ramm-Schmidt L. (1998) Bleach plant closure with latest evaporation technology. *Paper Asia*, **14** (11), 16-17.

Reith C. and Birkenhead B. (1998) Membranes enabling the affordable and cost effective reuse of wastewater as an alternative water source. *Desalination*, **117** (1-3), 203-209.

Rodrigo M. A., Michaud P. A., Duo I., Panizza M., Cerisola G. and Comninellis C. (2001) Oxidation of 4-chlorophenol at boron-doped diamond electrode for wastewater treatment. *Journal of the Electrochemical Society*, **148** (5), D60-D64.

Roorda J. (2001) Nuttig gebruik van RWZI-effluent. *H_2O*, **34** (3), 19-21.

Rose J. B., Friedman D. E. and Whalen T. (1999) The boiling effect : innovation for achieving sustainable clean water. *Water 21*, **1** (5), 16-19.

Routledge E. J. and Sumpter J. P. (1996) Estrogenic activity of surfactants and some of their degradation products assessed using a recombinant yeast screen. *Environmental Toxicology and Chemistry*, **15** (3), 241-248.

Rozzi A., Antonelli M. and Arcari M. (1999) Membrane treatment of secondary textile effluents for direct reuse. *Water Science and Technology*, **40** (4-5), 409-416.

Ruiters C., Kramer F., de Man A. and Hoeijmakers A. (1999) Effluent als bron voor industriewater in oostelijk Zuid-limburg. *H_2O*, **32** (23), 50-52.

Sacher F., Haist-Gulde B. and Brauch H.-J. (2001) Removal of drug residues during drinking water treatment. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Sander, R. (1999) *Compilation of Henry's law constants for inorganic and organic species of potential importance in environmental chemistry*. Max-planck institute of chemistry, http://www.mpch-mainz.mpg.de/~sander/res/henry.html, Mainz, Germany.

Saracco G., Solarino L., Specchia V. and Maja M. (2001) Electrolytic abatement of biorefractory organics by combining bulk and electrode oxidation processes. *Chemical Engineering Science*, **56** (4), 1571-1578.

Schafer A. I., Fane A. G. and Waite T. D. (2000) Fouling effects on rejection in the membrane filtration of natural waters. *Desalination*, **131** (1-3), 215-224.

Scholz M. and Martin R. J. (1997) Ecological equilibrium on biological activated carbon. *Water Research*, **31** (12), 2959-2968.

Schröder H. F. (2001) Mass spectrometric monitoring of the degradation and elimination efficiency for hardly eliminable and hardly biodegradable, polar compounds by membrane bioreactors. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Schwarzenbach R. P., Gschwend P. M. and Imboden D. M. (1993) *Environmental organic chemistry*. Wiley & Sons, New York.

Seo G. T. and Ohgaki S. (2001) Evaluation of refractory organic removal in combined biological powdered activated carbon - microfiltration for advanced wastewater treatment. *Water Science and Technology*, **43** (11), 67-74.

Seo G. T., Suzuki Y. and Ohgaki S. (1996) Biological powdered activated carbon (BPAC) microfiltration for wastewater reclamation and reuse. *Desalination*, **106** (1-3), 39-45.

Shah A. R. and Ploeser J. H. (1999) Reusing rinse wastewater at a semiconductor plant. *Journal of the American Water Works Association*, **91** (8), 58-65.

Shen Z. M., Wang W. H., Jia J. P., Ye J. C., Feng X. and Peng A. (2001) Degradation of dye solution by an activated carbon fibre electrode electrolysis system. *Journal of Hazardous Materials*, **84** (1), 107-116.

Shiklomanov I. A. (1993) World fresh water resources. *In: Water in crisis: a guide to the World's fresh water resources*. Gleick P. H. (ed.), Oxford University Press, New York, 13-24.

Simond O., Schaller V. and Comninellis C. (1997) Theoretical model for the anodic oxidation of organics on metal oxide electrodes. *Electrochimica Acta*, **42** (13-14), 2009-2012.

Sirotkin A. S., Koshkina L. Y. and Ippolitov K. G. (2001) The BAC-process for the treatment of wastewater containing non-iogenic synthetic surfactants. *Water Research*, **35** (13), 3265-3271.

Sozanski M. M. (1995) The effect of increase water salinity on the removal of micropollutants by biological carbon filters. *Environmental Technology*, **16** (8), 1061-1071.

Spencer P. (2000) Wastewater reuse and recovery: gain not pain. *Filtration & Separation*, **37** (1), 14-16.

Speth T. F., Gusses A. M. and Summers R. S. (2000) Evaluation of nanofiltration pre-treatments for flux loss control. *Desalination*, **130** (1), 31-44.

Squire D. (2000) Reverse osmosis concentrate disposal in the UK. *Desalination*, **132** (1-3), 47-54.

Squire D., Murrer J., Holden P. and Fitzpatrick C. (1996) Disposal of reverse osmosis membrane concentrate. *Desalination*, **108** (1-3), 143-147.

Standiford F. C. (1998) Evaporation. *In: Kirk-Othmer Encyclopaedia of Chemical Technology*. 4 th edition, John Wiley & Sons, New York, 959-981.

Stuetz R. M., Fenner R. A. and Engin G. (1999a) Assessment of odours from sewage treatment works by an electronic nose, H_2S analysis and olfactometry. *Water Research*, **33** (2), 453-461.

Stuetz R. M., Fenner R. A. and Engin G. (1999b) Characterisation of wastewater using an electronic nose. *Water Research*, **33** (2), 442-452.

Suzuki T., Watanabe Y., Ozawa G. and Ikeda S. (1998) Removal of soluble organics and manganese by a hybrid MF hollow fibre membrane system. *Desalination*, **117** (1-3), 119-129.

Tanghe T. (1999) *Biological and physico-chemical removal of xeno-estrogenic alkylphenols from aquatic environments*. Ph.D. thesis, Ghent University.

Tanghe T., Devriese G. and Verstraete W. (1999) Nonylphenol and estrogenic activity in aquatic environmental samples. *Journal of Environmental Quality*, **28** (2), 702-709.

Tchobanoglous G., Darby J., Bourgeous K., McArdle J., Genest P. and Tylla M. (1998) Ultrafiltration as an advanced tertiary treatment process for municipal wastewater. *Desalination*, **119** (1-3), 315-322.

Thiemann M., Scheibler E. and Wiegand K. W. (1987) Nitric acid, nitrous acid and nitrogen oxides. *In: Ullmann's Encyclopaedia of Industrial Chemistry*. Elvers B., Hawkins S. and Schulz G. (eds.), 5 th edition, VCH, Weinheim, 293-339.

Thoeye C., Geenens D., Vandaele S. and Van Houtte E. (2000) Need and technological possibilities of wastewater reuse in Flanders. *In: AWWA/WEF water reuse*, 30 January - 2 February 2000. AWWA, San Antonio, Texas.

Tleimat B. W. and Tleimat M. C. (1996) Water recovery from and volume reduction of gray water using an energy efficient evaporator. *Desalination*, **107** (2), 111-119.

Tsujimoto W., Kimura H., Izu T. and Irie T. (1998) Membrane filtration and pre-treatment by GAC. *Desalination*, **119** (1-3), 323-326.

US EPA (1992) *Guidelines for water reuse*. EPA/625/R-92/004, United States Environmental Protection Agency, Washington, D.C.

US EPA (1998) *Water recycling and reuse: the environmental benefits*. EPA/909/F-98/001, United States Environmental Protection Agency, Water Division Region IX, San Francisco, California.

US EPA (2001) *Removal of endocrine disruptor chemicals during drinking water treatment processes*. EPA/625/R-00/015, United States Environmental Protection Agency, Office of Research and Development, Washington, D.C.

van der Hoek J. P., Hofman J., Bonne P. A. C., Nederlof M. M. and Vrouwenvelder H. S. (2000) RO treatment: selection of a pre-treatment scheme based on fouling characteristics and operating conditions based on environmental impact. *Desalination*, **127** (1), 89-101.

van der Hoek J. P., Hofman J. and Graveland A. (1999) The use of biological activated carbon filtration for the removal of natural organic matter and organic micropollutants from water. *Water Science and Technology*, **40** (9), 257-264.

van der Roest H. F., van Bemtem A. G. M. and Lawrence D. P. (2001) MBR-technology in municipal wastewater treatment: challenging the traditional treatment technologies. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Van Hege K., Dewettinck T., Claeys T., De Smedt G. and Verstraete W. (2002) Reclamation of treated domestic wastewater using biological membrane assisted carbon filtration (BioMAC). *Accepted for publication in Environmental Technology*.

Van Hege K., Dewettinck T. and Verstraete W. (2001) Pre-evaporative Fenton remediation of treated municipal wastewater for reuse purposes. *Environmental Technology*, **22** (5), 523-532.

van Hoof S., Duyvesteijn C. and Vaal P. P. R. (1998) Dead-end ultrafiltration of pre-treated and untreated WWTP effluent for reuse in process water applications. *Desalination*, **118** (1-3), 249-254.

van Hoof S., Hashim A. and Kordes A. J. (1999) The effect of ultrafiltration as pre-treatment to reverse osmosis in wastewater reuse and seawater desalination applications. *Desalination*, **124** (1-3), 231-242.

Van Houtte E. (1999) Bereiding van drinkwater uit biologisch gezuiverd afvalwater met behulp van membraanfiltratie. *In: KVIV studiedag inzet membraantechnieken*, 5 mei 1999. KU Leuven, Heverlee.

Van Houtte E., Verbauwhede J., Vanlerberge F. and Cabooter J. (2000) Comparison between different out-to-in filtration MF/UF membranes for the re-use of biologically treated wastewater effluent. *In: Membrane technology in water and wastewater treatment*. Hillis P. (ed.), Royal society of chemistry, London, 190-197.

Van Houtte E., Verbauwhede J., Vanlerberghe F., Demunter S. and Cabooter J. (1998) Treating different types of raw water with micro- and ultrafiltration for further desalination using reverse osmosis. *Desalination*, **117** (1-3), 49-60.

Vandaele S., Geenens D. and Thoeye C. (2000) Integrated management of available water resources in Flanders (Belgium). *In: Proceedings of the 4th international Symposium on Biotechnology*, Hartmans S. and Lens P. (eds.), European Federation of Biotechnology, Noordwijkerhout, The Netherlands, 161-165.

Vandevivere P. C., Bianchi R. and Verstraete W. (1998) Treatment and reuse of wastewater from the textile wet-processing industry: Review of emerging technologies. *Journal of Chemical Technology and Biotechnology*, **72** (4), 289-302.

Vedavyasan C. V. (2000) Combating water shortages with innovative uses of membranes. *Desalination*, **132** (1-3), 345-347.

Venczel L. V., Arrowood M., Hurd M. and Sobsey M. D. (1997) Inactivation of *Cryptosporidium parvum* oocysts and *Clostridium perfringens* spores by a mixed-oxidant disinfectant and by free chlorine. *Applied and Environmental Microbiology*, **63** (4), 1598-1601.

Verhaege M., Verstraete W., Lissens G. and Van Hege K. (2002) Elimination of organic pollutants from plating rinse solutions by electrochemical treatment. *To be presented at: Recycling and waste treatment in mineral and metal processing - technical and economic aspects*, 16-20 June. TMS, Lulea, Sweden.

Virk G. S., Ford M. G., Denness B., Ridett A. and Hunter A. (2001) Ambient energy for low-cost water desalination. *Desalination*, **137** (1-3), 149-156.

Visvanathan C., Ben Aim R. and Parameshwaran K. (2000) Membrane separation bioreactors for wastewater treatment. *Critical Reviews in Environmental Science and Technology*, **30** (1), 1-48.

VLAREM II (2001) *Milieuwetboek Vlarem II*. Heyman J. and Smout L. (eds.), Kluwer rechtswetenschappen, Antwerpen.

Vlyssides A. G., Papaioannou D., Loizidoy M., Karlis P. K. and Zorpas A. A. (2000) Testing an electrochemical method for treatment of textile dye wastewater. *Waste Management*, **20** (7), 569-574.

Wade N. M. (2001) Distillation plant development and cost update. *Desalination*, **136** (1-3), 3-12.

Wang P., Lau I. W. C. and Fang H. H. P. (2001) Electrochemical oxidation of leachate pre-treated in an upflow anaerobic sludge blanket reactor. *Environmental Technology*, **22** (4), 373-381.

Wang S. (1998) Zero-liquid discharge possible using evaporation and crystallisation. *Water and Wastewater International*, **13** (5), 20-22.

Warta C. L., Papadimas S. P., Sorial G. A., Suidan M. T. and Speth T. F. (1995) The effect of molecular oxygen on the activated carbon adsorption of natural organic matter in Ohio river water. *Water Research*, **29** (2), 551-562.

Watanabe Y., Kimura K. and Suzuki T. (2000) Membrane application to water purification process in Japan - development of hybrid membrane system. *Water Science and Technology*, **41** (10-11), 9-16.

WCED (1987) *Our common future*. The World Commission on Environment and Development, Oxford University Press, Oxford, New York.

Weber W. J. and LeBoeuf E. J. (1999) Processes for advanced treatment of water. *Water Science and Technology*, **40** (4-5), 11-19.

WHO (1989) *Health guidelines for the use of wastewater in agriculture and aquaculture. Report of a WHO scientific group*. Technical report series 778, World health organisation, Geneva, Switzerland.

Wilf M. (1998) Reverse osmosis membranes for wastewater reclamation. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 263-344.

Witgens T., Gallenkemper M. and Melin T. (2001) Elimination of endocrine disrupting compounds from wastewater by membrane processes. *In: Proceedings of the 2nd world water congress of the International Water Association*, 15 - 19 October 2001. IWA, Berlin.

Wu W. W., Benjamin M. M. and Korshin G. V. (2001) Effects of thermal treatment on halogenated disinfection by-products in drinking water. *Water Research*, **35** (15), 3545-3550.

Yates M. V. and Gerba C. P. (1998) Microbial considerations in wastewater reclamation and reuse. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 437-489.

Young R. E., Thompson K. A., McVicker R. R., Diamond R. A., Gingras M. B., Ferguson D., Johanessen J., Herr G. K., Parsons J. J., Seyde V., Akiyoshi E., Hyde J., Kinner C. and Oldewage L. (1998) Irvine Ranch Water District's reuse today meets tomorrow's conservation demands. *In: Wastewater reclamation and reuse*. Asano T. (ed.), Technomic Publishing, Lancaster, 941-1036.

Zinkus G. A., Byers D. B. and Doerr W. W. (1998) Identify appropriate water reclamation technologies. *Chemical Engineering Progress*, **95** (5), 19-31.

CURRICULUM VITAE

Name	Kris Van Hege
Postal address	Hoogstraat 151 9550 Herzele Belgium
Tel.	+ 32 / (0) 486 / 53.21.36 (mob.) + 32 / (0) 53 / 62.42.39
E-mail	Kris.VanHege@rug.ac.be Kris.VanHege@c4.com
Place and date of birth	Zottegem September 6th, 1975
Nationality	Belgian
Marital status	Single

EDUCATION

1/1999-12/2001	Doctoral training in Applied Biological Sciences, Faculty of Agricultural and Applied Biological Sciences, Ghent University, Belgium
10/2000-4/2001	Post academic degree in ICT systems administration, Faculty of Engineering, Ghent University
10/1993-06/1998	Materials and metallurgical engineering, Faculty of Engineering, Ghent University, Belgium (distinction) Thesis: "Separation of Mo and Re from effluents generated by roastgas washinstallations"; Scientific promotor: Prof. Dr. M. Verhaege
09/1987-06/1993	Mathematics, Koninklijk Atheneum Zottegem (great distinction)

PROFESSIONAL RECORD

12/1998-12/2001	Doctoral research (doctoral scholarship) carried out at the Laboratory of Microbial Ecology and Technology (Ghent University) with prof. dr. ir. W. Verstraete as scientific promotor. Research was performed in collaboration with Aquafin N.V. and Severn Trent Water Ltd. Project: “Reuse of treated effluent of domestic wastewater treatment plants in industry”

WORK EXPERIENCES

12/1998-12/2001	Co-ordinator of research projects commissioned by Milliken, Bonduelle, Procter & Gamble
12/1998-07/2001	Tutor of 5 students during their Master’s thesis
12/1998-1/2001	Network administrator of the local computer network (35 computers)
12/1998-10/2000	Responsible for computational exercises of the courses Microbial Ecological Processes and Biotechnological Processes of Environmental Sanitation
7/1997-8/1997	Research project carried out at the Council for Scientific and Industrial Research (CSIR), Pretoria, South Africa
7/1996-8/1996	Research project carried out at the Council for Scientific and Industrial Research (CSIR) in co-operation with Pretoria Portland Cement (PPC), Midrand, South Africa

PUBLICATIONS

Verhaege M., Van Hege K. and Wettinck E. (1999) Supported liquid membranes: a feasible technology for the treatment of effluents containing refractory metals. *In: Global symposium on recycling, waste treatment and clean technology*, 5-9 September, San Sebastian, Spain.

Dewettinck T., Van Hege K., Tanghe T. and Verstraete W. (1999) Doorgedreven zuivering van afvalwater voor hergebruik in industriële processen. *Annales Medicinae Militaris Belgicae*, **13** (1-2), 43-48.

Dewettinck T., Van Hege K. and Verstraete W. (2001) The electronic nose as a rapid sensor for volatile compounds in treated domestic wastewater. *Water Research*, **35** (10), 2475-2483.

Dewettinck T., Van Houtte E., Geenens D., Van Hege K. and Verstraete W. (2001) HACCP to guarantee safe water reuse and drinking water production - A case study. *Water Science and Technology*, **43** (12), 31-38.

Van Hege K., Dewettinck T. and Verstraete W. (2001) Pre-evaporative Fenton remediation of treated municipal wastewater for reuse purposes. *Environmental Technology*, **22** (5), 523-532.

Dewettinck T., Van Hege K. and Verstraete W. (2001) Development of a rapid pH-based biosensor to monitor and control the hygienic quality of reclaimed domestic wastewater. *Applied Microbiology and Biotechnology*, **56** (5-6), 809-815.

Dewettinck T., Hulsbosch W., Van Hege K., Top E. M. and Verstraete W. (2001) Molecular fingerprinting of bacterial populations in groundwater and bottled mineral water. *Applied Microbiology and Biotechnology*, **57** (3), 412-418.

Van Hege K., Dewettinck T., Claeys T., De Smedt G. and Verstraete W. (2002) Reclamation of treated domestic wastewater using biological membrane assisted carbon filtration (BioMAC). *Environmental Technology, accepted*.

Van Hege K., Verhaege M. and Verstraete W. (2002) Indirect electrochemical oxidation of reverse osmosis membrane concentrates at boron-doped diamond electrodes. *Electrochemistry Communications*, **4** (4), 296-300.

Verhaege M., Lissens G., Van Hege K. and Verstraete W. (2002) Elimination of organic pollutants from plating rinse solutions by electrochemical treatment. *Paper to be presented in: Recycling and waste treatment in mineral and metal processing: technical and economic aspects*, 16-20 June. TMS, Lulea, Sweden.

Hammes F., Séka A., Van Hege K., Van De Wiele T., Vanderdeelen J. and Verstraete W. (2002) Calcium removal from industrial wastewater by biocatalytic $CaCO_3$ precipitation. *Submitted*.

Van Hege K., Verhaege M. and Verstraete W. (2002) Electro-oxidative abatement of reverse osmosis membrane concentrates. *Submitted*.

PARTICIPATION TO CONGRESSES, SEMINARS AND WORKSHOPS

1999 Seminar Berson UV techniques, Nuenen, Nederland, February 10

Workshop “Hergebruik” (Aquafin), KVIV Antwerp, February 24

Workshop “Mens en water”, Military hospital Queen Astrid, Brussels, October 22

“Binnenkijken bij bedrijven”, Seminar within the framework of the Euregio Scheldemond project (GOM West-Vlaanderen), December 7

Oral Presentation: “Treatment of WWTP effluent for reuse in industrial processes”

2000 BIWA happy hour, seminar of the Belgian branch of the International Water Association, Brussels, March 14

Poster presentation: “Advanced treatment of WWTP effluent for reuse in industrial processes”

Milieucontactnamiddag “Grondwaterwinning en rationeel watergebruik” (GOM Oost-Vlaanderen), Ghent, May 11

BIWA happy hour, seminar of the Belgian branch of the International Water Association, Brussels, June 13

1st World water congress of the International Water association, Paris, July 3-7

14th Forum for Applied Biotechnology (FAB), Bruges, September 27-28

2001	BIWA happy hour, seminar of the Belgian branch of the International Water Association, Brussels, February 19
	15th Forum for Applied Biotechnology (FAB), Ghent, September 24-25
	7th Faculty of Agricultural and Applied Biological Sciences PhD-symposium, Ghent, October 10. *Poster presentation: Biological membrane assisted carbon filtration for reuse of wastewater treatment plant effluent*
	2nd World Water congress of the International Water Association (IWA), Berlin, October 15-19 *Poster presentation: Assessment of the fouling potential of wastewater plant effluent using a modified capillary suction time measurement*
	BIWA happy hour, seminar of the Belgian branch of the International Water Association, Brussels, October 22
	Scientec Matrix 2001 International Symposium on Catalytic Water Treatment, Antwerp, November 25-27 *Poster presentation: Biological membrane assisted carbon filtration for reuse of wastewater treatment plant effluent*