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Emerging contaminants and treatment options in water recycling for indirect potable use

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ABSTRACT

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

Key words | emerging contaminants, indirect potable reuse, water recycling

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INTRODUCTION

Population growth, increased industrial activities in many parts of the world as well as the estimated impacts of climate change will exceed various pressures on the water supply systems (EEA 2007). Particularly in developing countries there is a need to dramatically enlarge the access to safe and piped water (UN 2006). Considering the growing burden on freshwater resources, particularly the over-abstraction of groundwater, the deterioration of groundwater quality, and saltwater intrusion into coastal aquifers, the utilisation of alternative water sources is a promising option to supplement water supply and restore natural resources (EEA 1999; Margat & Vallée 2000). Among the alternative water sources are seawater, brackish groundwater, urban storm water, collected rain water as well as reclaimed water which can be used for different beneficial purposes.

Global water demand will continue to increase and is expected to reach probably 70% of the available freshwater resources till 2025 (UN 2006). Furthermore, these resources are unequally distributed and often of poor quality so that numerous regions in the world face rising water stress. Superposed on the evolution of demand are natural and man-made variations of water availability.

The water stress index – the ratio of a country's total water withdrawal to its total renewable freshwater resources – serves as a rough indicator for the pressure exerted on water resources. Not all water uses are causing comparable stress. A ratio in the range of 10% to 20% indicates that water availability is becoming a constraint on development. A water stress index above 20% is supposed to necessitate comprehensive water management efforts and actions to resolve conflicts among competing uses (OECD 2003). It is striking that approximately half of the European countries, representing almost 70% of the population, are already facing water stress issues today.

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Water reclamation and reuse applications

Municipal wastewater including stormwater can be considered as an alternative water source if treated appropriately for the intended use. This is highlighted by the Urban Wastewater Treatment Directive (91/271/EEC) and by the Water Framework Directive (2000/60/EC) encouraging "reuse measures" and "artificial recharge" as supplementary measures that can be applied to reach the fixed environmental objectives for surface and groundwater bodies.

Reclamation and reuse of municipal wastewater is a method to mitigate increasingly evident water stress arising in many regions of the world from water scarcity and water quality degradation. The proposed implementation of water reclamation in integrated water resources management is driven by sustainability issues, potential cost-advantages and climate change conditions. The socio-cultural environment often drives and determines the appropriateness of technical solutions (Bixio *et al.* 2005; Asano *et al.* 2006).

Indirect potable reuse

Indirect potable reuse can be regarded as any augmentation of water bodies utilised for drinking water supplies by wastewater treatment plant effluents or overflows from combined sewer systems (Asano *et al.* 2006). One can distinguish between unintended or *de facto* indirect potable reuse which happens along major river catchments around the world, where drinking water resources are influenced by wastewater discharges as observed on basis of anthropogenic tracer substances, (Ternes & Joss 2006; Weil & Knepper 2006; Dillon & Jiminez 2007) as well as planned or intended indirect potable reuse. Planned indirect potable reuse is a measure to supplement public water supply by the utilisation of reclaimed water through introduction in a drinking water reservoir or via managed aquifer recharge.

Managed recharge has for a long time provided means to mitigate depletion of groundwater levels, to protect coastal aquifers from saltwater intrusion, and to store surface water for future use (Kanarek & Michail 1996; Mills *et al.* 1998; Bouwer 2000). Aquifer storage has some advantages over surface water reservoirs which might be more costly and environmentally questionable. Moreover, soil percolation and aquifer storage act as treatment steps while avoiding evaporation as well as taste and odour problems due to algae growth in surface storage (Dillon 2000; Asano & Cotruvo 2004). Recharge can be performed in two ways: direct or indirect. If conducted by infiltration and percolation through soil and subsoil, the recharge processes, e.g. by so-called Soil Aquifer Treatment (SAT), offer an additional barrier. Mixing reclaimed wastewater with natural groundwater prior to any intended use also positively influences the public acceptance of a reuse scheme.

The concept of Aquifer Recharge (AR) offers potential for various subsequent uses like irrigation, industrial process water and augmentation of public water supplies. The latter, indirect potable use, is certainly one of the most challenging water reclamation and reuse applications with a high demand in terms of safety because of the potential use as drinking water (whose quality criteria are fixed in directive 98/83/EC) and the general level of protection required for groundwater resources as laid down in the directive on the protection of groundwater against pollution and deterioration (2006/118/EC). Even if drinking water supplementation is not explicitly foreseen in an aquifer recharge project, the provision of drinking water quality in the recovered product is a common bottom line in many applications, e.g. in Israel where the Dan Region Reclamation and SAT scheme provides "accidental drinking water quality" on a large scale (Mekorot 2003).

EMERGING CONTAMINANTS IN INDIRECT POTABLE USE

Water quality criteria

According to a recent review conducted by Asano & Cotruvo (2004) the main concerns in indirect potable reuse are health risks resulting from pathogens and trace chemicals as well as nitrate. Among the organic pollutants there is a range of so called emerging contaminants such as endocrine disrupting compounds (EDC), pharmaceutically active compounds (PhAC), personal care products (PCP) and disinfection by-products (DBP). The occurrence of these emerging contaminants in the aquatic environment including surface and groundwaters, and wastewater sludges as well as drinking water has been studied (Heberer 2002; Ternes &

Table 1 | Emerging contaminants in indirect potable reuse and aquifer recharge schemes

Scheme and sample point	Compound	Concentration [ng/L]	Reference
North City Water Reclamation Plant San Diego (CA) – Advanced Water Treatment Pilot			
◆ Tertiary effluent	Sulfamethaxoazole	892	DeCarolis *et al.* 2006
◆ Reverse osmosis permeate	Sulfamethaxoazole	2.9	DeCarolis *et al.* 2006
◆ UV + Peroxide product	Sulfamethaxoazole	< 1.0	DeCarolis *et al.* 2006
◆ Tertiary effluent	Iopromide	632	DeCarolis *et al.* 2006
◆ Reverse osmosis permeate	Iopromide	1.4	DeCarolis *et al.* 2006
◆ UV + Peroxide product	Iopromide	< 1.0	DeCarolis *et al.* 2006
Bolivar Aquifer Storage and Recovery Scheme (South Australia)			
◆ Reclaimed water	Atrazine	9.2	Overacre *et al.* 2006
◆ Groundwater (5 month storage)	Atrazine	4.5	Overacre *et al.* 2006
◆ Groundwater (11 month storage)	Atrazine	1.8	Overacre *et al.* 2006
◆ Reclaimed water	Estrone	32	Overacre *et al.* 2006
◆ Groundwater (5 month storage)	Estrone	24	Overacre *et al.* 2006
◆ Groundwater (11 month storage)	Estrone	11	Overacre *et al.* 2006
Temporary Hanningfield reservoir augmentation by Chelmsford effluent (UK)			
◆ Sewage plant effluent	Estrone	8 33	Gomes & Lester 2003
◆ UV-treated effluent	Estrone	< 1-20	Gomes & Lester 2003
Infiltration of secondary effluent into groundwater via a wetland upstream of a drinking water facility (Germany)			
◆ Receiving surface water	Carbamazepin	1,100	Bergman *et al.* 2003
◆ Wetland	Carbamazepin	500–600	Bergman *et al.* 2003
◆ Groundwater (420 m from infiltration)	Carbamazepin	530	Bergman *et al.* 2003
Irrigation of treated wastewater in Braunschweig, Germany			
◆ Tertiary effluent	Sulfamethaxoazole	620 ± 90	Ternes *et al.* 2007
◆ Groundwater (in the irrigation area)	Sulfamethaxoazole	< LOQ – 110	Ternes *et al.* 2007
◆ Tertiary effluent	Carbamazepine	2,100 ± 700	Ternes *et al.* 2007
◆ Groundwater (in the irrigation area)	Carbamazepine	< LOQ – 570	Ternes *et al.* 2007
◆ Tertiary effluent	Diatrizoate	1,700 ± 3,300	Ternes *et al.* 2007
◆ Groundwater	Diatrizoate	1,600 – 9,600	Ternes *et al.* 2007

Table 1 | (continued)

Scheme and sample point	Compound	Concentration [ng/L]	Reference
Comparing MF, RO and soil-aquifer treatment for indirect potable reuse of water (USA)			
◆ Tertiary effluent	EDTA	35,400 ± 27,600	Drewes *et al.* 2003
◆ Microfiltration permeate	EDTA	5,600	Drewes *et al.* 2003
◆ Nanofiltration permeate	EDTA	< LOQ	Drewes *et al.* 2003
◆ Reverse osmosis permeate	EDTA	< LOQ	Drewes *et al.* 2003
◆ Soil-aquifer treatment	EDTA	3,960 ± 3,180	Drewes *et al.* 2003
◆ Tertiary effluent	APECs	55,800 ± 59,400	Drewes *et al.* 2003
◆ Microfiltration permeate	APECs	23,400	Drewes *et al.* 2003
◆ Nanofiltration permeate	APECs	< LOQ	Drewes *et al.* 2003
◆ Reverse osmosis permeate	APECs	< LOQ	Drewes *et al.* 2003
◆ Soil-aquifer treatment	APECs	680 ± 1,140	Drewes *et al.* 2003
Water samples influenced by wastewater in Berlin (Germany)			
◆ Surface water	p-TSA	<1,150	Richter *et al.* 2007
◆ Groundwater (directly below a former sewage farm)	p-TSA	<40,800	Richter *et al.* 2007
◆ Drinking water	p-TSA	<270	Richter *et al.* 2007
◆ Surface water	BSA	<520	Richter *et al.* 2007
◆ Groundwater (directly below a former sewage farm)	BSA	<1,220	Richter *et al.* 2007
◆ Drinking water	BSA	<50	Richter *et al.* 2007

LOQ = limit of quantification, p-TSA = paratoluenesulfonamide, BSA = benzenesulfonamide, EDTA = ethylenediaminetetraacetic acid, APECs = alkylphenolpolyethoxycarboxylates.

Joss 2006). A particular substance of concern is N-nitrosodimethylamine (NDMA) which was found to be a possible by-product in water and wastewater disinfection (Mitch & Sedlak 2002) and is characterised as highly carcinogenic.

In contrast to the wide range of EDCs, PhACs and PCPs, only DBPs are currently considered in water related pieces of regulation and supra-national recommendations, particularly in drinking water quality requirements such as the WHO Guidelines for Drinking-water Quality (WHO 2004), the European Drinking Water Directive and the US National Primary Drinking Water Regulations (US EPA 2006). Guidelines for indirect potable use make regularly reference to drinking water quality standards and add a number of specific criteria such as absence of faecal contamination and low TOC (>3 mg/L) (US EPA 2004; Salgot *et al.* 2006). While not formally incorporated into

Table 2 | Water reclamation schemes investigated in RECLAIM WATER (www.reclaim-water.org)

Site location & capacity	Scheme description
Sabadell 30 km from Barcelona, Spain Capacity: 25,000 $m^3 d^{-1}$	Secondary treated wastewater effluent discharged into a river bed where it infiltrates and is recovered. The water is then disinfected (UV) and distributed for parks irrigation.
Nardò Salento Region, south of Bari, Italy Capacity: 12,000 $m^3 d^{-1}$	Secondary treated municipal effluent is transported to aquifer injection. Recharge acts as a salt intrusion barrier and resource is also used as drinking water source.
Shafdan Negev, Israel Capacity: 342,000 $m^3 d^{-1}$	Secondary wastewater from the Tel-Aviv area is recharged to an aquifer via a soil aquifer treatment (SAT) system. Recovered water is primarily used for irrigation but has accidental drinking water quality.
Gaobeidian Beijing, China Capacity: 650 $m^3 d^{-1}$	Tertiary effluent is used for aquifer recharge. Treatment is provided by coagulation, filtration and ozonation (in test) prior to infiltration and recharge.
Adelaide Salisbury, South Australia Capacity: 1,100 $m^3 d^{-1}$	Wetland treated urban stormwater injected into a brackish aquifer. Water recovered via separate recovery wells. Recovered water intended for drinking supplies, and until proven will be used for irrigation.
Torrele (Wulpen) Belgium Capacity: 8,640 m^3/d Infiltration: 2.5 million m^3/ year	Tertiary treated municipal effluent is upgraded by microfiltration and reverse osmosis, and then infiltrated via an infiltration pond to prevent salt intrusion and to recharge an aquifer used for drinking water production.
Mezquital Valley (State of Mexico) Mexico Infiltration: 630 million m^3/year	Wastewater mixed with stormwater and surface water is discharged to an irrigated area of more than 76,000 ha. About 40% of the irrigation water infiltrates into the aquifer. The water is recovered via separate wells and springs. 206 well systems, 31 springs, and 63 waterwheels are in operation. Recovered water is chlorinated and distributed for drinking water supply, industrial use, irrigation and other purposes (bathing, swimming, washing).
Atlantis (Cape Town) South Africa Capacity: 15,000 m^3/d Infiltration: 2.7 million m^3/year	Urban stormwater run-off, is collected via a series of detention basins and blended with secondary treated domestic wastewater and recharged up-gradient of the production well field for augmenting the water supply. The blend of natural groundwater and recharged water abstracted from the well field is used as potable water supply for the town of Atlantis.
NEWATER Singapore Capacity: 150,000 m^3/d	NEWATER is treated used water further purified using dual-membrane (microfiltration and reverse osmosis) and UV treatment. Four NEWATER factories are in operation supplementing Singapore's water supplies, part of the product water is use to augment drinking water reservoirs.

national drinking water standards the Californian Office of Environmental Health Hazard Assessment issued 2006 a Public Health Goal (PHG) for NDMA of 3 ng/L (OEHHA 2006). The California Department of Health Services' Division of Drinking Water and Environmental Management (DDWEM) has established a notification level of 10 ng/L for a number of nitrosamines (DHS 2006). Without a direct link to indirect potable use, but to guideline values for emerging contaminants in the aquatic environment, the European Parliament Committee on the Environment, Public Health and Food Safety has approved a proposal in 2007 for an extension of the list of the priority substances to be included in the Annex X of the Water Framework Directive (EPC 2007), containing trace organics as bisphenol A, carbamazepine, clotrimazole and diclofenac.

Occurrence and fate of emerging contaminants

The occurrence and fate of emerging contaminants has been monitored in a number of indirect potable reuse and aquifer recharge schemes (PUB 2002; Gomes & Lester 2003; Asano *et al.* 2006; Snyder *et al.* 2007). Table 1 summarises selected data on the occurrence and fate of emerging contaminants obtained from monitoring of both planned and unplanned

Table 3 | Nanofiltration application studies in water and wastewater applications

Application	Treatment aims	Source
Drinking water treatment (Groundwater)	Water softening	Gorenflo *et al.* (2002)
Drinking water treatment (Surface water)	TOC removal	Beros *et al.* (2003)
Textile industry	Dye removal	Van der Bruggen *et al.* (2004)
Desalination	Salt removal	Melin & Rautenbach (2007)
Water reclamation	TOC removal	Meier & Melin (2005)

indirect potable reuse as well as aquifer recharge schemes. To overcome the limitations of compound-specific tests with respect to the potential health hazards toxicological tests have also been carried out with bio-assays in a number of indirect potable reuse schemes. Both toxicological tests as well as a limited number of epidemiological tests could not show that a higher health risk is connected to water recycling than to the use of the conventional sources considered (PUB 2002; Khan & Roser 2007)

TREATMENT OPTIONS IN INDIRECT POTABLE REUSE

Wastewater is generally more polluted than most drinking water sources and hence requires more extensive treatment prior to indirect potable use. The treatment technology for these ambitious goals is generally available and implemented in full scale (e.g. in Orange County, Singapore and Wulpen/Belgium). Examples exist where the aquifer is recharged with secondary treated effluent only and the soil-aquifer system is used as treatment and storage, as it is the case in economically developed countries such as Israel, USA and Australia (International Water and Irrigation Review 1999) as well as in countries with transient economies such as Mexico, South Africa, Thailand and Peru (Dillon & Jiminez 2007). The natural attenuation processes occurring in the soil and sub-soil, particularly in the vadose zone, have shown to be quite effective with respect to trace organic removal (Ternes *et al.* 2007). On the other hand, in the last decade the common practice in Western Countries is rather to apply tertiary (e.g. coagulation, filtration, disinfection) or quaternary treatment (various types of double membrane systems, as it is the case in Belgium, California, Singapore, Australia and Namibia) before infiltration and potable use (Bixio *et al.* 2005).

The effectiveness of a range of intensive water reclamation technologies such as conventional and membrane coupled (MBR) activated sludge treatment, membrane effluent filtration with porous and dense membrane processes, activated carbon adsorption as well as different oxidation processes (Ozone, UV + Ozone, UV + Peroxide) have been investigated in a number of studies (Asano *et al.* 2006; Ternes & Joss 2006; Snyder *et al.* 2007). It seems obvious from the result of these investigations that an almost complete retention of a wide range of emerging contaminants can be achieved with multi-barrier treatment processes. Particularly difficult is the removal of NDMA; advanced oxidation processes with UV and H_2O_2 have been implemented in indirect potable reuse schemes in the US to cope with this problem (Asano *et al.* 2006).

Technologies applied in full scale water reuse schemes

There is a number of treatment technologies applied in indirect potable reuse projects ranging from soil application of raw wastewater to highly engineered double membrane systems plus advanced oxidation. Compilations of the different treatment trains utilised in various schemes worldwide can be obtained e.g. from Asano *et al.* (2006), Ternes & Joss (2006), Dillon & Jimenez (2007). A number of different indirect potable reuse and managed groundwater recharge schemes are under investigation by the European Project partnership RECLAIM WATER. The case studies investigated there also with respect to the occurrence and fate of trace organics are listed in Table 2.

Table 4 | Retention of organic micropollutants with NF and RO membranes

Compounds	Substance type	Membrane type	Retention [%]	Source
Bisphenol A	EDC	NF	1.9–99.7*	Gallenkemper (2005)
Bisphenol A	EDC	RO	18–83	Kimura *et al.* (2004)
Bisphenol A	EDC	NF	47– > 99	Agenson *et al.* (2003)
Nonylphenol	EDC	NF	90.5–99.3*	Gallenkemper (2005)
Estrone	EDC	NF/RO	13– > 80	Nghiem *et al.* (2004)
Estrone	EDC	NF	80– >95	Schäfer *et al.* (2003)
Estrone	EDC	NF	65–83	Braeken *et al.* (2005)
Estrone	EDC	NF	40– > 99	Weber (2004)
Estradiol	EDC	NF	20– > 80	Nghiem *et al.* (2004)
Estradiol	EDC	NF	49– > 99	Weber (2004)
Estradiol	EDC	RO	29–83	Kimura *et al.* (2004)
Diethylstilbestrol	EDC	NF	60– > 99	Weber (2004)
Mestranol	EDC	NF	90– > 99	Weber (2004)
Ethinylestradiol	EDC	NF	41– > 99	Weber (2004)
Atrazine	Pesticide	NF	59–92	Zhang *et al.* (2004)
Atrazine	Pesticide	NF	86–95	Chen *et al.* (2004)
Atrazine	Pesticide	NF	> 80	Plakas *et al.* (2006)
Simazine	Pesticide	NF	45– > 80	Zhang *et al.* (2004)
Primidone	Pharmaceutical	RO	85–87	Kimura *et al.* (2004)
Isopropylantipyrine	Pharmaceutical	RO	69–78	Kimura *et al.* (2004)
Carbamazipine	Pharmaceutical	RO	85–91	Kimura *et al.* (2004)
Sufamethoxazole	Pharmaceutical	RO	70–82	Kimura *et al.* (2004)

*retentions are corrected by the effect of concentration polarisation.

Emerging technologies

Among the emerging technologies for indirect potable use nanofiltration should be highlighted as a treatment technology which can remove a wide range of microbiological as well as chemical contaminants (Schäfer *et al.* 2005). Nanofiltration can be considered as an alternative to reverse osmosis technology where a lower degree of desalination is required. With a molecular weight cut-off above 200 g/mol it is a promising treatment option for a variety of emerging trace contaminants. Nanofiltration has been investigated for a number of purification applications as shown in Table 3.

The largest full scale implementation of nanofiltration in water treatment applications is the Méry sur Oise plant close to Paris that was built to reduce the high organic load in the raw surface water utilised for drinking water production. Seasonal pesticide issues were the reason to complement the multi-barrier concept by a dense membrane process (Cyna *et al.* 2002; Beros *et al.* 2003).

In a number of studies the organic micropollutant removal capacity of nanofiltration has been investigated and compared to reverse osmosis (Table 4). Medium to high removal rates have been observed for endocrine disrupting compounds, pesticides and pharmaceutically active compounds.

In a recent study conducted in Germany on the application of dense membrane processes to treat River Rhine bank filtrate reverse osmosis was preferred when compared to nanofiltration due to the even higher micropollutant retention detected in this case (Loi-Brügger *et al.* 2007).

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