REVIEWING THE APPLICATION OF MULTI-BARRIER MEMBRANE FILTRATION TECHNOLOGIES TO RECLAIM MUNICIPAL WASTEWATER FOR INDUSTRIAL WATER USE

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ABSTRACT

The significant percentage of the world's water consumption devoted to industrial use, along with an increasingly higher environmental concern of society, have awaken the interest of industry on using municipal reclaimed water for replacing fresh water use coming from utilities or natural resources. Depending on the type of industry and the specific application, water must meet certain quality requirements. Therefore, those water quality standards that are required for those most relevant industrial applications wherein the use of reclaimed water has noticeably been reported are herewith reviewed. Although the use of internal water treatments for recycling and reusing their own effluents has recently and widely been reported within many industrial sectors worldwide, the substitution of fresh water by reclaimed municipal wastewater has not been much extended yet. The increasing proportion of municipal wastewater reclamation plants that rely on membrane filtration technologies versus the total number of reclamation facilities that are worldwide distributed is also assessed within this review, including the discussion of their main related drawbacks.

*Keywords***:** membrane filtration, municipal wastewater reclamation, industrial water quality, ultrafiltration, reverse osmosis.

1. INTRODUCTION

Water shortage, an increasing population, and a more stringent legislation regarding water conservation and environmental compliance, are imposing severe restrictions to industrial water use since its activity entails about a quarter of the world's total water consumption (1). Moreover, predictions inferred from climate change considerations may even suppose more stringent water use policies in the short term because a significant impact is expected on water resources quality and availability (2).

Although water recycling within industrial facilities leads to reduce water consumption figures, it is very difficult to achieve a total closure of their water circuits because residual contaminants eventually accumulate in reused water (3), and fresh water intake is always somewhat needed to compensate losses (e.g. by evaporation) in any case (4). As a consequence, and despite industrial process-water consumption is progressively being reduced as a result of a higher closure level of water circuits, the demand for this resource will still be large in the close future. Fortunately, the remaining water demand after closing water circuits might alternatively be satisfied by reclaimed water from municipal sewage treatment plants in many cases.

Despite its great application potential, the market for reclaimed water is nowadays still awaiting a wider exploitation. The major factors limiting the use of reclaimed water in industry are: availability to ensure continuous operation without water shortage, meeting quality standards, volume, cost, and reliability (5, 6). In short, reclaimed water use only satisfied about the 0.4% of the total water use worldwide demanded for industrial purposes in 1995 (7); whereas this alternative only reached the 1% ten years later (2005) (8). Agriculture is the main user of reclaimed water (70%), followed by industry (20%), and domestic uses (10%) (9).

Beyond these average figures, there are strong significant differences among how the volume of reclaimed wastewater is distributed in each country. For example, Tunisia devotes an 86% water use for agricultural purposes, for which a 30-45% of the treated sewage is also used (10). On the other hand, USA, Singapore, and Germany, respectively devote a 45%, 51% and 69% of their fresh water capacity to industrial purposes, and many recycling and reuse projects have therefore been implemented within their industry (9). Finally, only about the 10% of the fresh water resource of Spain is consumed by industrial applications (11). Correspondingly, only a 0.3% of the Spanish reclaimed sewage capacity was consumed by industry in 2009 (≈1 hm^3/year) (12); although an enterprising plan aiming to boost the use of reclaimed water in all sectors is currently being implemented, estimating an accumulated industrial use of 38 hm^3/year (6.2%) at a 2015 horizon (12).

In tune with these considerations, there is much available information regarding municipal wastewater reclamation initiatives for irrigation purposes in agriculture or environmental applications (13-15); as well as for implementing treatments to reclaim the effluent within specific industrial facilities (16-18). However, the use of reclaimed municipal wastewater for industrial applications is still much less extended; with the exception of cooling systems, where municipal reclaimed water has been used from long time ago after a conventional tertiary treatment (19). In addition, reclaimed municipal water has been also used in several other industrial auxiliary applications, significantly replacing fresh water intake, such as washing and flushing, and in general cleaning and watering plants (7, 8).

Nevertheless, the interest in using reclaimed sewage as industrial process water is expected to increase in the close future, provided its economical (4, 20-21) and constant quality features (21), and new initiatives being developed and reported within several industrial sectors (4, 20, 22, 23). Particularly, it may represent a very interesting alternative for industrial facilities located within areas suffering water scarcity; furthermore when authorities (24) and climate change forecast (2) are progressively imposing more stringent limitations to fresh water consumption. In short, many companies may also find that the substitution of fresh water by municipal reclaimed water may be an economically feasible complement to compensate water losses within their own closed circuits, and/or a good alternative to building on-site specific treatments for recycling their own effluents (4, 23).

In general terms, guidelines defining water quality parameters that reclaimed water must fulfil for its different potential use alternatives follow two main orientations: (a) those who prefer limiting health hazard to the maximum extent possible, being the "*Californian Title 22*" (24) regulation a major example; and (b) those who pretend to be more realistic and feasible, such as the World Health Organisation's "*Guidelines for the safe use of wastewater and excreta in agriculture and aquaculture*" (1989) (10). In short, most countries (even states or regions within) have developed their own guidelines regarding the use of reclaimed water combining quality standards from both legislation types, so there are no universal water quality standards defined (20).

Nevertheless, although every state has its own legislation within the USA, all of them are collected within the "*Guidelines for Water Reuse*" (25). Correspondingly, some Australian jurisdictions have together implemented the "*Australian Guidelines for Water Recycling: Managing Health and Environmental Risks*" (26); as well as they have developed specific validation requirements for recycled water schemes (27). On the other hand, there are no current formal definitions or guidelines for water reuse at the European level. Conversely, there are very different water reuse practices from North to South, and even among Mediterranean countries (28). In fact, the European

Union is at least making a big effort to solve this legislation disagreement creating the "*Mediterranean-EU Water Initiative Working Group for Wastewater Reuse*", which is first one collecting information regarding the current status of wastewater reuse within the EU and the Mediterranean area in order to implement specific demand-driven actions (29). Within this framework, "*Aquarec*" EU's project has defined seven water quality categories including microbial and chemical limits for each one (30).

Despite water quality guidelines for industrial use differ among countries and the standards required for each specific application, health hazard is the main issue that is always mentioned within all available legislations because special attention should be paid when workers might be in direct contact with water. In this sense, both "*Californian Title 22*" and the Australian guidelines define a minimal water reclamation process based on a secondary treatment followed by coagulation-flocculation, filtration or flotation, and disinfection. On the other hand, they differ in the limits related to the allowed presence levels of microorganisms. For example, "*Californian Title 22*" establishes a total coliforms threshold of <23 colony-forming units (CFU)/100 mL for specific sampling (<2.2 weekly) (24); whereas the Australian standards set <10 CFU/100 mL for *E. Coli* (26); but both legislations agree limiting turbidity to <2 NTU.

Beyond these considerations, current reclamation treatment trends are focused on obtaining the highest water polishing grade in order to avoid any health hazard or operational risk. In this sense, membrane processes (microfiltration, MF; ultrafiltration, UF; nanofiltration, NF; and reverse osmosis, RO) are taking competitive advantage over those physico-chemical processes specified in historically used standards within municipal wastewater treatment plants (mWWTP) (9). In short, membrane processes produce high quality water regardless the variations in the characteristics of the inflowing feed water in comparison to other treatment processes (31). Particularly, MF and UF are applied as the preferred processes for the retention of microbial and suspended solids; and as pre-treatments for NF or RO stages, which are able to generate process water with a very high quality standard, even drinking water (32, 33).

Industrial water use is herein reviewed considering water quality requirements for different uses, and the possibility of using municipal reclaimed water as an alternative to fresh water use. Municipal wastewater reclamation facilities that produce treated water for industrial purposes are also located worldwide providing an overall water reclamation assessment. In particular, membrane filtration alternatives, including their potential drawbacks, for reclaiming municipal wastewater for industrial purposes are also reviewed, provided these technologies are currently taking the lead within the alternatives to polish sewage.

2. ASSESSING CURRENT AND POTENTIAL APPLICATIONS FOR RECLAIMED MUNICIPAL WASTEWATER IN INDUSTRY

Most standards for water use are set to maintain a proper operation of industrial systems, to preserve the quality of manufactured products, and to protect public and environmental health. Main water quality guidelines that are applicable to major industrial water consumers are reviewed next.

2.1 Utilities

2.1.1 Cooling water

More than 50% of the fresh water consumed by industry is used for cooling (34). Power generation plants, petroleum refineries, chemical manufacturers, metal processors, and food (i.e. mainly meat and dairy products) and beverage manufacturers, are the major consumers of cooling water. In 2004, power generators from the USA accounted for approximately the 40% of the freshwater withdrawal within the country (35). Although this value finally only accounts for about the 3% of the total freshwater consumption in the USA (the rest 37% is returned back), it still represents a significant demand of freshwater use (36). The United States Environmental Protection Agency (USEPA) established the cooling tower water quality requirements within their "*Guidelines for water reuse*" (EPA/625/R-92/004), published in 1992 (37) (Table 1), which have not been modified so far.

Figure 1 shows water use intensity for different fuel types and cooling systems in thermoelectric power plants where cooling towers are typically placed (38). In short, the amount of required water depends on the type of cooling system and the efficiency of the turbine, which increases as the difference between steam and condensation temperatures increases as well. Therefore, more efficient plants require less cooling effort to produce the same amount of energy. For example, coal plants nowadays operate at higher temperatures than nuclear ones, so they consume less water.

In recirculating cooling systems, such as cooling towers or spray ponds, water evaporates as it dissipates process heat, so the water solution become concentrated many times (typically 4-8 times) (39). As a consequence of both an elevated concentration of diverse matter and such high water temperature, scaling, corrosion and biofouling trouble may be promoted in the system. Particularly, ammonia is a critical compound that may form metal complexes affecting metals solubility and release, as well as it may also promote biological growth and bio-corrosion. In fact, it is especially corrosive to copper alloys at a concentration greater than 2 mgN/L (40). In addition, silica may precipitate when its concentration threshold favours silicic acid polymerization until the solubility limit of amorphous silica is reached, which is further decreased by the presence of trace amounts of aluminum (41); as well as iron content is able to deactivate polymers that are used to inhibit calcium phosphate scaling (421). Finally, organic matter content thresholds that may induce the formation of metal complexes, which may further increase metal release as well, or promote biological growth and consequent bio-corrosion trouble in cooling systems, may be controlled monitoring total organic carbon (TOC) concentration in the solution (1).

Moreover, cooling water systems (particularly open recirculating ones such as cooling towers) provide a favourable environment (i.e. T= 25-45 ºC; pH= 5.5-8.5) for the growth of bacteria, algae, fungi, protozoa, and viruses (43-44); being the gram negative aerobic bacteria *Legionella pneumophila* especially dangerous for public health, as it is the responsible to cause the fatal infectious *Legionellosis* disease. All these microorganisms can be drifted into the environment through cooling equipment aerosols causing other diverse illnesses (45-49). In short, all of them may enter the system in makeup water, through the air, or from process leaks; and they will then begin to proliferate if an effective preventive monitoring and maintenance programme is not implemented. Finally, microbial growth on wetted surfaces ultimately leads to the formation of biofilms, which are made up by the accumulation of microorganisms themselves, extra-cellular secreted products, and other organic and inorganic debris. These biofilms affect equipment performance adversely and also promote metal corrosion (43, 50-51).

The key to limit bio-growth, and the consequent slime formation, in any water system reside in controlling nutrients contents circulating in water streams. Besides nitrogen- and phosphorous-based compounds, nutrients may be provided by scale (Ca and Mg based), suspended solids, corrosion products, or trapped organic or inorganic molecules supplied by the incoming water (43). Although corrosion and scaling are relatively easy to control in cooling systems keeping water chemical contents under the values reported in Table 1 (37), corrosion inhibitors and sequestrating agents are normally added to their water circuits. In order to avoid biological contamination, it is really important to initiate an appropriate biocide treatment program at system's start-up; and continue thereafter on a regular basis following the instructions received from the supplier of the implemented biocide treatment (52).

2.1.2 Boiler-feed water

Water quality requirements for boilers are clearly more restrictive than those for cooling water (Table 1) (37) due to the higher temperatures and pressures that are required for steam production; particularly when the heat source is in direct contact with the tubes. As boiler-feed water quality requirements depend on the applied pressure, more restrictive water quality figures are in general required at higher pressures (38).

In short, those critical quality parameters that have been associated to boiler water use are alkalinity, and silica, iron, manganese, and copper (37, 38). Two operational problems have been associated to high alkalinity figures: (a) as water temperature increases, CO_2 is released increasing corrosion potential; and (b) carbonate can contribute to foaming, which leads to deposit formation in the superheater, reheater, and/or turbines (1). In addition, silica can form scale if pressure is below 600 psig. Above this threshold, silica starts to volatize, passing over with steam to potentially form deposits on the steam turbine diaphragms and blades (53). Finally, the most common type of galvanic corrosion that is found in boiler systems is that caused by the contact of dissimilar metals, such as iron and copper (53).

2.1.3 Sealing water

The amount of water required for sealing vacuum pumps may represent a high fresh water consumption within common industrial processes. For example, the total sealing water quantity that is used in the vacuum pumps of a paper mill may sum up to 5-10 m^3 per tonne of paper produced (54).

Those quality requirements that have been reported for sealing water are also comparatively listed in Table 1 (55). The most restrictive parameter to consider is the proportion of abrasive matter content. In short, if solids are incorporated as water flows, they may cause premature seal failure and, ultimately, damage to the pumps. Temperature and salts content must be also carefully controlled, provided the precise role of sealing water is keeping the rotor system cool, and particularly considering that scaling is susceptible to occur inside the system.

2.2 Process water

The alternative use of reclaimed water in industrial processes mainly depends on its final application (Table 1). Electronic and pharmaceutical industrial processes, for example, require process water of almost distilled quality for some production applications. In fact, four different water quality standards are typically defined for pharmaceutical industrial use: potable water, purified water, highly purified water (HPW) and water for injections (WFI). In short, HPW requirements are shown in Table 1 (56). In addition, food processing operations are a bit less restrictive, only requiring drinking water quality when there is direct process water contact with the products that are going to be consumed by people. On the other hand, leather-tanning industrial processes require relatively low-quality water use. Textile, pulp and paper, and metal industrial sectors usually demand intermediate requirements. The quality standards that are required for process water within those main industrial sectors using reclaimed water for their applications are considered next.

2.2.1 Pulp and paper industry

Fresh water is mainly used as dispersion and transport media for fibrous raw materials and additives along the production process developed in paper mills, from pulping to forming. In addition, water is also required as heat exchanging fluid, sealant in vacuum systems, lubricant agent, for the production of steam, and in high and low pressure showers (55-57).

At the beginning of the 20th century, water consumption in the pulp and paper sector was estimated to be over 600 m^3 per tonne of pulp produced. Nowadays, depending on the type of final paper product, raw material used, and the optimization of water use (including the installation of internal kidneys), modern paper mills just consume 4-100 m^3 per tonne (58-60). In short, Figure 2 shows maximum and minimum fresh water consumption values in modern paper mills manufacturing different types of paper (55). Actual consumption figures mainly depend on the water circuit optimization level that has been achieved within each paper mill.

Depending on the specific application where it is going to be used and the paper grade being produced, process water must meet certain chemical and physical criteria (Table 1) (61); which can be complemented by an even more restrictive set of values if recommended by industrial equipment suppliers. In short, the control of hardness and alkalinity is required to avoid the formation of calcium aggregates with organic colloids in the pulp suspension, and scaling in machinery and process circuits. In fact, the presence of colloidal material may produce losses in paper resistance and quality. Moreover, it could block showers, affecting the quality of the final product as well. In addition, some metals (Fe, Al, or Mn), chlorides, and sulphates, are highly corrosive; the latter may also produce scaling and odour problems (62-63).

2.2.2 Textile industry

The typical processing operations of the textile sector (e.g. washing, scouring, dyeing, printing, bleaching and finishing) require a great variety of water consumption thresholds. In short, it has been quantified that wool and felted fabrics processes are larger water consumers than other fabrics such as woven, knits, stock and carpet (Figure 2) (64-68). Moreover, the amount of water used within the same fabric processing subcategory varies within a wide range depending on the achieved level of water circuit closure and the efficiency of the mill.

It would be pretty difficult defining a general quality standard for reclaimed water to be used within textile mills because water quality characteristics that are required within this industry are strongly linked to the type of fibre being processed (silk, cotton, polyester, etc.), the textile process itself (scouring, desizing, dyeing, washing, etc.), and the quality of the fabric produced. In general, turbidity and colour are undesirable for many textile purposes (Table 1) (5). Moreover, whether colour is produced by the presence of some organic compounds, it may exert demand for chlorine in the bleaching stage, and therefore reduce the effectiveness of chlorine as bleaching agent. In addition, suspended solids may also include organisms that could cause spots, or degradation of dressing and/or finishing solutions (66).

Furthermore, iron salts could be harmful to textile processing in many ways (69). In scouring and bleaching stages, they may provide yellow tinge to white material. In dyeing stages, iron may combine with some dyes and cause dulling effects. On the other hand, other metal ions may also act as catalysts in the decomposition of bleaching agents (70). Finally, hardness may cause deposits on textiles causing problems in those processes using soap (71, 72). All these basic parameters that must be monitored when considering the use of reclaimed water in processes within textile industry are shown in Table 1 (5).

2.2.3 Food industry

Food processing plants need water for many purposes: washing and transporting raw materials and products, cleaning installations and equipments, thawing, canning, cooling, steam generation, etc. (73). Figure 3 shows a summary of the reported water consumption volumes for the majority of food, drink, and milk (FDM) sectors (74). Dairy sector particularly, followed by fish processing mills, and deodorization processes, account for those significant maximum water consumption figures.

As water may be in contact with food in some processes, the use of reclaimed water for food industrial applications may raise certain controversy in many countries. For example, the use of reclaimed water for food processing applications is currently forbidden under Spanish and Italian legislations (75-76). In general terms, the contact between food and reclaimed water is not allowed; as well as water quality must meet the standards of drinkable water in any other case. In this sense, the minimum drinking water requirements that are called for within the European Union are described in the Council Directive 98/83/EC (77), which defines water quality standards for human consumption. Correspondingly, all water contacting food should meet the standards set by their "*Drinking Water Guidelines*" in Australia (78); and there is a similar situation in the USA, where only potable water quality is allowed for cooking or being added to food products, as well as the quality of the final product must not be affected (i.e. water must be free of dissolved minerals that could make water excessively hard or affect taste somehow) (73, 79).

2.2.4 Electronic industry

Electronic microchip manufacturing requires a large volume of ultrapure water (UPW); as this is the primary cleaning agent that is used in silicon wafer production processes (80). In short, UPW is mainly required for rinsing circuits, channels, and gates in order to remove chemicals that have been added along the manufacturing process. In fact, inorganic salts, and organic matter that may remain or be deposited on the wafer surface during processing, directly affect the number of usable devices that may be produced from a single silicon wafer (80); so rinsing with high purity water can therefore significantly increase electronic microchip production. Since water contacts wafer surface several times during manufacturing, its quality demand must be the highest.

For example, a typical 200 mm-wafer fabrication plant that produces 40,000 units per month uses as much water as 50,000 people would consume in a month (80). A survey developed in 2005 for Hsin-Chu Science-based Industrial Park (HSIP, Taiwan) showed that 0.79, 4.0, and 7.3 m^3 of water were necessary to produce one single 6-in, 8-in, and 12-in wafer, respectively (81). In addition, 0.2266 m^3 per wafer were reported to be required for the fabrication of one LED crystal; 0.025 m^3 per wafer were addressed for a single nickel-plated aluminium plate; and 0.94 L per set were necessary to produce a wireless card (81).

In general, the water quality standards that are demanded within the electronic industrial sector are discretionarily set by the owner of each production plant; and they are not specifically regulated by any public or private organism besides the existent general legislation in rule regarding health hazard and environmental concern in each country. Therefore, each manufacturing operation develops an internal quality specifications demand from its own processing requirements, or from sources of standards and general specifications such as the American Society for Testing and

Materials (ASTM), Semiconductor Equipment and Materials International (SEMI), Balazs Labs, or Sematech. Particularly, ASTM D5127-99 corresponds to the "*Standard Guide for Ultrapure Water Used in the Electronics and Semiconductor Industry*" (82). In short, general reference specifications for water quality within the electronic industry are collected in Table 1 as well (83).

2.2.5 Chemical industry

It has been recently reported that the chemical industry is consuming the 11% of the total water withdrawal in Europe (84), which approximately represents the 3% of the water directly abstracted by all industry (85). Nevertheless, the general trend is showing a significant reduction of these values; that is, the European Chemical Industry Council (CEFIC), for example, has reported that chemical industrial facilities have reduced their water consumption about an 8% from 2003 to 2007.

Water quality requirements and consumption figures widely differ among chemical industrial applications, provided this sector includes such a great diversity of manufactured products (e.g. basic chemicals, steel, fuel, petrochemicals, coal, other mining products, plastics, detergents and toiletries, medicines, paint, pesticides, etc.) For example, the total amount of water that is used in oil refineries has been estimated as 0.25-0.34 m^3 of water per crude oil barrel (86); and extracting 1 barrel of bitumen from oil sand takes 2 to 3 barrels of water (87). In short, Table 1 also summarizes USEPA's general guidelines for process water quality demands in the chemical industry, and the basic quality standards that are required in the petrochemical and coal sectors (88).

3. MEMBRANE TECHNOLOGIES FOR WATER RECLAMATION

Membrane filtration is basically based on placing a selective barrier between two phases.

As a result of exerting a driving force to one side of the membrane, components are transported towards the membrane surface. Therefore, some components pass through the membrane (permeate) and others are retained according to their size (retentate). Considering a particular industrial application, different membrane systems and configurations might be arranged, including pre-treatments and other treatment stages based upon different technologies, in order to meet those target water quality standards that this industrial application may specifically demand from reclaimed municipal wastewater.

3.1. Types of membrane systems available for wastewater reclamation

In general terms, membrane technologies may be classified according to the nature of the applied driving force (i.e. type of pressure or vacuum), or by the operating size ranges for filtration. In short: *microfiltration* (MF) can separate 0.1-10 μm particles from solvents or other low-molecular components, for which an operating pressure of 0.1-2 bar is required; *ultrafiltration* (UF) is applied to retain macromolecules or sub-micrometrical particles (0.002-0.1 μm), requiring operating pressures of 0.1-5 bar; *nanofiltration* (NF) is very useful for removing micro-pollutants, herbicides, and bivalent ions (Ca^{2+}, Mg^{2+}, SO_4^{2-}, CO_3^{2-}) due to its 0.5-2 nm pore-size filtration range, for which a 3-20 bar pressure is necessary; and *reverse osmosis* (RO) is able to separate macromolecules and low molecular mass compounds (>1-0.1 nm), such as monovalent ions (Na^+, K^+, Cl^-, NO^{3-}, etc.) and sugars, working at higher pressure thresholds (5-120 bar) in order to overcome higher osmotic pressure values in the solution (89).

A complete membrane treatment unit comprises membranes (in assembled modules), a pressure support structure, a feed inlet, a concentrate outlet, and an overall support structure. The main types of *membrane modules* that are used for wastewater treatment are: *plate-and-frame*, *pleated cartridge*, *tubular, capillary*, *hollow fibre* and *spiral wound*. Each type of module shows specific advantages and disadvantages depending on feed water quality, their design, and maintenance protocols (90); whereas main differences among them are related to the allowed filtration area and their capacity to promote turbulence to reduce fouling occurrence.

In addition, manufacturers offer membrane modules made of a wide variety of materials (e.g. polypropylene, polysulfone, polyvinylidene difluoride, polyether-sulfone, cellulose acetate, aromatic polyamide, regenerated cellulose, titanium oxide, etc.) showing different physico-chemical behaviours (e.g. mechanical strength, oxidant tolerance, pH operating range, etc.). In short, their compatibility to each specific operational set of conditions should be verified by end-users (89-91).

The direction of feed-water flow in relation to the surface of the membrane determines the type of filtration in MF and UF systems. For example, an *encased membrane system* may use a *cross-flow design*, where part of the feed stream is withdrawn as permeate, whereas the other part is forced to flow across the membrane surface; or it may operate in *dead-end mode*, where the total feed water volume passes through the membrane, leaving all components that are larger than its pores in the feed's phase. This option results effective when the concentration of particles in the feed stream is low (turbidity <10 NTU), or the packing tendency of the filtered material does not produce large pressure drop across the membrane (90). On the other hand, water streams characterized by a higher concentration of particles or macromolecules (i.e. 10-100 NTU) will rapidly compact the cake if operated in dead-end mode, so an unacceptable quick pressure drop will be driven (92).

Similarly, semi-permeable membranes of NF and RO systems do not allow the passage of organics or salts, so dead-end filtration will result in plugging or fouling(90);

although NF filtration may be operated at both modes. In this sense, cross-flow systems are the best alternatives to keep stable filtration rates; despite they consume more energy because their pumping systems have to guarantee high superficial velocities (1-6 m/s) through a constant recirculation loop (90, 93, 94).

Summing up, although any membrane design can be applied for wastewater with a low suspended solids content, only specifically designed membranes and operation modes would be able to handle effluents carrying high amounts of solids. Particularly, higher cross-flow velocities would be required in these cases. In short, it is highly recommended to perform optimization trials whenever a specific membrane filtration system is designed and implemented for reclaiming a specific wastewater quality.

Besides an encased design, membrane modules can be arranged conforming a *submerged membrane system* inside a tank containing wastewater, where filtration is performed by the application of vacuum. A lower sensitivity to fouling and a lower filtration pressure requirement are the main advantages of submerged systems; whereas some disadvantages can be also addressed, namely: i) foam episodes; ii) fouling may be boosted by high recovery rates, as a higher discharge frequency is required; iii) a higher potential health hazard as a consequence of the exposition of workers to wastewater contact; and iv) the operation of bubbling agitation (which facilitates filtration and reduces fouling) has to be balanced with its operational cost (4).

In addition, the direction and orientation of feed water across the surface of the membrane define inside-out and outside-in operation modes. Feed water surrounds the membrane in *outside-in* systems, so filtrate is collected inside the hollow fibre (lumen). In an *inside-out* system, feed water enters to the fibres at one end of the membrane element, and discharge stream passes through the element exiting at the opposite side. In this system, filtrated water is collected inside the element on the outside of the fibres.

In general, inside-out systems are more sensitive to fouling; therefore requiring more intense wastewater pre-treatment, and more frequent cleaning (95).

Finally, *membrane bioreactors* (MBRs) represent a particular configuration of submerged systems, where biological degradation of waste products is integrated with membrane filtration (96). As a consequence, the separation of solids does not depend on settling, and a smaller reactor volume is needed. In addition, they may be used with high filamentous bacteria contents and floating sludge, allowing a very long retention time for solids and generating much less sludge (96-99). In addition, MBR effluents are suitable for feeding RO polishing stages without installing additional pre-treatment processes in between. On the other hand, MBRs are relatively expensive to install and operate; as well as they address other limitations imposed by a low oxygen transfer efficiency (due to a high organic presence), pressure, temperature, and pH requirements (96-99).

There are two main MBR configurations: 1) *submerged membranes* (*S-MBR*), where the filtration unit is integrated inside the bioreactor; and 2) *side-stream MBR* (external circulation), where a separate membrane module recycles the rejected stream back to the bioreactor. As permeate flux is relatively low in S-MBR configurations, it can be held for extended periods of time without decline; but membrane permeability could suffer rapid losses if solids content is high (>20 g/L), as well as an important membrane fouling may be generated at low mean cell residence times (≅ 2 days) (98). On the other hand, both flux and fouling rates are much higher in side-stream MBRs; as well as they require more frequent, and rather more aggressive, cleaning operations, which can be properly performed backwashing or adding chemicals.

Summing up both operational and cost issues, it has been reported that submerged configurations operate more effectively than side-stream ones when they are applied to reclaim domestic wastewater, considering both energy consumption and cleaning requirements (99). Specifically, aeration results to represent the main operating cost component of these systems because it is required for both mixing and transferring oxygen. In addition, S-MBRs operate at lower flux regimes requiring a greater membrane area, which also implies a greater associated investment. In particular, it has been demonstrated that a side-stream UF-MBR system fed with urban wastewater may produce reclaimed water of enough quality for municipal watering, toilet flushing, and car washing. Moreover, it could be also used for cooling; even as process water after softening (100).

3.2. Challenges to improve the application of membrane filtration to reclaim municipal wastewater

Although membrane filtration generally produce high grade quality water requiring low footprint, and its implementation is nowadays following an increasing trend for municipal wastewater reclamation, they must also face some drawbacks and challenges for a future wider application.

3.2.1 Fouling and cleaning

It is widely known that fouling is the main cause for membrane's loss of performance because it leads to flux decline, trans-membrane pressure increase, higher energy requirements, membrane biodegradation, and a greater salt passage rate (101, 102). Several types of membrane fouling have been identified, including those caused by suspended solids and particles, colloids, scaling, metal oxidation, organics, extracellular polymeric substances (EPS), and biological organisms (102-105) In fact, membranes are usually affected by one or two different types of fouling at the same time, which are summarized in Table 2 (104, 106-109).

In general, *dissolved organic matter* (effluent organic matter, EfOM) typically present in municipal wastewater (TOC ≈ 5-20 mg/L; BOD_5 ≈ 3-10 mg/L) (110), is composed of a wide range of constituents (polysaccharides, proteins, aminosugars, nucleic acids, humic and fulvic acids, organic acids, EPS, and cell components). Together to other colloidal matter is able to partially pass (depending on the pore size) through MF or UF membranes causing important fouling to those subsequent NF or RO membranes that are disposed in serial (111). In particular, polysaccharides have been found to play the most important role in fouling NF and UF membranes (112); as well as the presence of soluble EPS (biomass supernatant) has been widely reported to play the main role in the formation of biofouling on MBR's membranes (109).

Although effluents from mWWTPs contain much lower total dissolved solids contents than seawater (1,500 versus 38,000 mg/L, respectively), *scaling* may also cause special trouble in RO membrane systems (113). Rivers, groundwater, rainfall, seawater, and municipal wastewater, contain basic or acidic inorganic species that are in equilibrium with precipitates, or with other potential scaling components, mainly: OH^-, F^-, CO_3^{2-}, SO_4^{2-}, PO_4^{3-} and $Si(OH)_4$; whereas cationic species that (directly or indirectly) are more likely to form precipitates in water include: Ca^{2+}, Mg^{2+}, Fe^{3+} and Al^{3+}. All these inorganic species are responsible to form those main precipitates (phosphates, sulphates, carbonates, hydroxides, and fluorides) that have been identified to cause scaling in reclaiming municipal wastewater RO systems (108).

Hydrophobicity, surface charge, pore size, and roughness, are *membrane properties* that have been related to greater fouling occurrence. In fact, it has been reported that if there are no crevices in the surface of the membrane, no material will be deposited there because the boundary layer formation becomes less pronounced, and cross-flow will be then able to remove it at a faster rate than it could be deposited (114).

In addition, the effect of *membrane charge* is mainly reflected by its role in controlling electrostatic adsorption and pore clogging. Particles populating wastewater are generally negatively charged due to the presence of organic matter that came from every source of water that made it up (original drinking water, human activity, surface run-off, etc.), as well as from the bacterial activity that may have been developed along sewage transport (115, 116). Therefore, membranes showing a neutral to negative net surface charge tend to show hydrophilic interaction or electrostatic repulsion with these constituents, further reducing biofouling episodes (117, 118). Finally, other water quality properties, such as pH value or its ionic strength, may also influence organic matter deposition (119). For example, these compounds usually show carboxyl ($-COOH$) or phenolic ($-OH$) functional groups, which further increase the concentration of negative charges as the pH value turns higher (120).

The diversity of *materials* that may be used to manufacture commercial UF/MF membranes span the range from fully hydrophilic polymers (e.g. cellulose acetate, CA) to fully hydrophobic ones (e.g. polypropylene, PP; and polyethylene, PE). Between both well-defined groups of polymers, the polysulfone (PS) - polyethersulfone (PES) family, polyacrylontrile (PAN), and polyvinylidene fluoride (PVDF) show an intermediate behaviour; and they may be blended with additives and pore formers in order to design moderate hydrophilic membranes (121). Particularly, sulfone (e.g. PS and PES) polymers are characterized by their mechanical strength, thermal and chemical stability, and excellent film forming properties; as well as their hydrophilic character, and their consequent *biofouling resistance* as well (122), may be easily increased blending them with other polymers thanks to their high inherent solubility. As a result, they may even achieve CA membranes hydrophilicity figures (123-124).

Considering all these properties favouring or preventing membrane fouling, much research effort has been recently devoted to modify membrane surface properties in order to further enhance their *anti-fouling behaviour* (125-127). Particularly, a double repulsive NF hollow fiber membrane, with a positively charged selective layer and a negatively charged substrate, has recently been developed with the ability to effectively retain diverse dye molecules contained in textile industrial wastewater over a wide range of pH values (127).

Another common strategy to *control fouling* relies on installing aeration devices in MF, UF, or even in some NF, systems in order to enhance surface membrane shear and therefore reduce foulant layer thickness. Bubbling can be applied within membrane modules, or externally to them in submerged systems; its effectiveness has been reported to be the same (128). In contrast, aeration may represent the 70% of the total energy cost of the treatment, so current research efforts aim to optimize this operation and its associated cost figures. In fact, it has already been reported that only the 10% of the actual operational time is optimally devoted to perform this task (129). Finally, enhancing shear rate via mechanical means seems it would be a potential alternative to bubbling as a mean for fouling control, as it has been shown by magnetically induced membrane vibration in MBRs (130).

Furthermore, fouling can be classified as reversible or irreversible depending on the effectiveness of both, fouling controlling strategies and selected cleaning technologies. *Reversible fouling* can be described as the fraction that can be effectively removed by one or a combination of the following methods: 1) flow inversion (backwashing or permeate backflushing); 2) forward flush (cross-flow at high flow rates); and 3) membrane surface scouring using relaxation breaks like it has been reported in MBR systems (131), or just bubbling air (128), among other alternatives.

In short, flux decline caused by irreversible fouling cannot be recovered unless membranes are cleaned by chemical agents, or ultimately replaced (90). Particularly, NF and RO membranes do not allow backwashing, so chemical treatment (namely the *clean-in-place* (CIP) operation) is the only feasible alternative for them (4, 90). The selection of those chemicals that are going to be used in CIP operation mainly depends on the type of foulant; although they generally belong to one of the following six categories: acids (e.g. HCl, HNO_3, H_2SO_4, H_3PO_4, oxalic acid, and citric acid), caustics (e.g. NaOH, KOH, and NH_4OH), sequestering complexes (e.g. EDTA), detergents (e.g. alkyl sulphate, and sodium dodecyl sulphate), enzymatic chemicals (e.g. alpha-CT, CP-T, and perozidase), oxidizing disinfectants (e.g. NaOCl, H_2O_2, and $KMnO_4$), and commercial blends (e.g. 4Aquaclean, Divos, Triclean, and Ultrasil/Aquaclean) (132, 133).

In addition, cleaning effectiveness may be enhanced combining *physico-chemical cleaning* methods, which are those mechanical cleaning processes (i.e. backwashing) that are assisted by the addition of certain chemical agents (i.e. NaOCl or citric acid). For example, a chemically enhanced cleaning operation that is generally applied for cleaning MBRs daily consist on performing backwash using permeate to which certain chemical products have been added in small quantities (109).

Summing up, the selection of the best *cleaning strategy* (type of chemical, cleaning conditions, and frequency) for backwashes and CIP is the key to achieve both, a constant membrane system performance, and the lowest possible contribution to operational cost (4, 134). While mechanical cleaning is directly related to the operational cost of membrane treatment because energy consumption is mainly derived from air supply; CIP step shows certain controversy because it requires long operation time, chemicals are consumed, and some membranes may be degraded (135, 136); as well as it may cause corrosion in the system, and it could result harmful to the environment when waste streams are discharged (137).

Current research on this topic focuses enhancing cleaning efficiency and reducing all the above mentioned drawbacks. Particularly, an "*enhanced flux maintenance*" (EFM) strategy has been recently postulated advantageous versus traditional CIP operation (138). In short, EFM firstly consists on flushing/soaking, or recirculating, the chemical solution to the membrane for a short period of time. The chemical solution is thereafter drained, and the membrane is ultimately flushed with feed water. As a result, an automated EFM strategy only requires 30-40 min to be completed, whereas CIP typically lasts several hours. Savings up about the 30% in the lifetime cost of membrane systems, as well as a 20% smaller footprint (a lower number of membranes is required), have already been reported for EFM (138).

Although an *enzymatic cleaning* is especially effective for protein-fouled membranes, and despite it is considered environmentally friendly, its associated cost is still prohibitive for large-scale applications; so *chemical-free cleaning strategies* (mechanical, mainly) are taking competitive advantage nowadays. Particularly, a new concept of *mechanical cleaning* (without chemicals) has been recently developed in MBR systems based on the use of a granular material that is introduced inside the membrane tank holding activated sludge. The operation mode consists on performing a constant filtration, without backwash and relaxation, where the granules touch or beat the surface of the membranes; therefore producing the desired cleaning effect (139). Moreover, it was confirmed that this application enhanced fluxes about a 20%, whereas the presence of the granules did not have a negative impact on the behaviour of the membrane (140). In addition, it has also recently been demonstrated that prepared *inert salt solutions* (e.g. $NaCl$, $NaNO_3$, Na_2SO_4, KCl, $CsCl$, and NH_4Cl), seawater, or brines from desalination plants, can be used as effective cleaning agents for organic fouling in RO systems because they are able to react with foulants through ion-exchange processes (141).

Furthermore, *sonication* cleaning protocols have ultimately been assessed in both MBRs (109) and NF systems (142). In short, this process is essentially based on breaking the fouling cake down into smaller fragments. However, it may not be effective for those fouling types causing blockage of the pores. Nevertheless, its combination with other cleaning processes may surely lead to achieve higher flux recovery figures. Finally, the application of *shear-enhanced filtration systems* has been also assessed as an alternative membrane cleaning process. Particularly, the use of a *magnetically induce membrane vibration system* has been reported to achieve promising results at lab-scale MBRs (130).

3.2.2 The management of rejects

Another significant problem that arises when operating membrane processes (NF and RO systems, mainly) for water reclamation in mWWTPs is the production of a concentrated stream (or brine) that is characterized by a high concentration of micropollutants, including: organic matter, refractory chemicals (e.g. pesticides, personal care products, and endocrine disruptors), residuals from wastewater treatment itself (e.g. soluble microbial products, partially biodegraded organics, and anti-scaling chemicals), and biological materials (e.g. bacteria, viruses, oocysts, and cell fragments) (143). These substances can negatively affect the environment if they are freely disposed without appropriate treatment (144). Whether UF and MF systems also produce rejected streams, they are not constantly being generated along operation; they are tightly linked to backwash frequency.

Although the diversity of harmful compounds that are contained within these rejected streams is pretty well-known, discharging them to surface water, oceans, and groundwater (depending on availability), has mainly been used as their predominant management alternative (145, 146). Halophyte irrigation has also been considered (144), but only when the original water source is not wastewater. Therefore, the development of alternative technologies that may allow recycling and reusing municipal wastewater within industrial processes, or in other applications, is highly encouraged under a more environmentally-friendly wastewater management perspective.

Different initiatives regarding the removal of hazardous components from concentrated streams have been reported to date in order to increase their quality standard before disposal. For example, advanced oxidation processes (AOPs), such as ozonation (147-149), Fenton processes, and UV/TiO_2 (150), have been demonstrated to be very efficient in the oxidation of a wide variety of pollutants typically present in retentated streams; as well as they may result even more efficient if a biodegradation stage is thereafter implemented (150).

In addition, electrolysis, followed by oxidation with hypochlorite, resulted successful to remove total ammonia nitrogen, COD, and colour, from RO concentrated effluents flowing out a mixed domestic and textile wastewater treatment plant (1519). Furthermore, denitrification has also successfully been reported using a bioactive fluidized bed adsorber reactor (152).

Biological methods (e.g. single sequential batch reactor and sequential anaerobic-aerobic two-sludge system) have been also reported capable to decolorize and reduce

COD values of the retentate produced by NF membranes used to polish textile effluents, for example (153).

Particularly, both, adsorption onto granular ferric hydroxide, and chemical precipitation with NaOH, were reported to be effective removing phosphate from NF concentrate (154). In addition, phosphate removal has been also assessed using polymeric ligand exchange resins that are able to recover phosphate forming struvite ($MgNH_4PO_4 \cdot 6H_2O$), which may be finally used as a fertilizer (155).

Nevertheless, none of these treatments would totally solve the problem. Although some compounds are removed, others equally dangerous remain, especially when the original stream is sewage. For example, evaporating retentate in ponds requires a large surface and, although the installation of multi-effect evaporators reduces footprint, it could finally result very expensive (156).

In short, the best management alternative for these streams would be finding direct applications for them, namely recycle them. For example, the successful use of membrane filtration retentate from a paper mill effluent for brick manufacturing has been recently reported (157). Finally, some preliminary studies have demonstrated that bipolar membrane electrodialysis and electrochlorination are suitable treatments for generating mixed acid and base streams, and some chlorine generation, respectively; which can be directly applied to RO systems, as well as in other upstream pre-treatments (158).

4. WORLDWIDE DISTRIBUTION OF FACILITIES RECLAIMING MUNICIPAL WASTEWATER FOR INDUSTRIAL APPLICATIONS

Freshwater scarcity for human consumption and agricultural purposes may be significantly reduced in the close future as current freshwater intake would be substituted by reclaimed municipal wastewater within those main industrial sectors which water quality requirements have previously been reviewed. Nevertheless, the number of facilities that nowadays supply municipal reclaimed sewage for industrial applications worldwide is still low (Figure 4) compared to the amount of them dedicated to produce reclaimed water for agriculture or urban purposes (10). In addition, important differences could be observed among different countries, continents even.

For example, reclaimed water is becoming of great importance in *Australia*, where legislation and standards impose that a minimum water use of the 20% should be supplied by reused water by 2012 (159). This requirement has represented a real driving force for industries and municipalities, encouraging them to actively develop research on water reuse and recycling. All municipal facilities serving reclaimed water for industrial purposes that have been found located in this country are based on multi-barrier membrane systems, namely MF/UF+RO (Table 3), and are mainly concentrated in the seaside area of Queensland and New South Wales (160-165). Only one membrane installation was found in Western Australia, Perth specifically (Figure 4). Sewage reclamation is not well extended yet in this area maybe because it has the highest amount of distributed water (≈30%) originated from groundwater resources (166), and their increased water demand has mainly been afforded by desalination facilities (167).

Two main water reclamation cores for industrial application can be clearly distinguished in *Asia*, namely *Singapore and China* (Figure 4 and Table 4). In order to reduce the amount of freshwater being imported from Malaysia, the government of *Singapore* has strongly promoted water reclamation projects. As a result, Singapore is the world's current leader implementing wastewater reclamation plants, including the creation of the first NEWater facilities in 2003, *Bedok* and *Kranji* plants (23- 168). NEWater is the brand name given to reclaimed water being produced by the

Singapore's Public Utilities Board (PUB). This reclaimed water has undergone very stringent purification and treatment processes using advanced dual-membrane systems (MF/RO or UF/RO) and ultraviolet disinfection technologies (Table 4). NEWater represents the 30% of Singapore's total water supply nowadays (169), in part because it has been demonstrated that NEWater costs half the price for desalinated water (170-171). Seven wastewater reclamation facilities have currently been located in Singapore (Figure 4); five of them producing NEWater (Table 4), which is not only used as process water for industrial purposes, but also serves as drinking water.

Wastewater reclamation is also especially important in North-Eastern *China* (Figure 4), where severe water shortage is emerging as a result of both a large population demand and increasing water pollution levels (172). However, municipal wastewater reclamation still needs a wider development within this country in order to increase its recycle rate (water recycled/water discharged) from the 2.7% achieved in 2007 up to the aimed 20% in 2015 (173). These figures particularly contrast Israel's 75% water recycling rate (174). Nevertheless, China's 11th five-year plan (2006-2010) clearly aimed to improve water reclamation infrastructure by awarding new projects and upgrading facilities that were already in operation (175). As a result, most water reclamation facilities that have been found located in China (Figure 4) were started-up during this period of time *(*176); and those implementing membrane filtration are the same in number, although their capacity is lower, than those applying other treatments based on the "*California Title 22*" (24, 176).

Europe locates just a few municipal wastewater reclamation facilities devoted to supply reclaimed water for industrial applications (Figure 4). In addition, all of them are of small capacity (Table 5); and main reuse projects in the Mediterranean region are related to agricultural, landscape irrigation, or groundwater recharge requirements (177).

Particularly, those plants located in Peterborough (UK) and Madrid (Spain) are based on multi-barrier membrane systems. The first one (MF+RO) started up in 2002, and it produces 1,200 m^3/day of permeate for steam generation at Peterborough's Power Station (178). Madrid's one (UF+RO) is currently being built inside *"Cuenca Media-Alta de Arroyo Culebro"* mWWTP, and it will provide more than 10,000 m^3/day of reclaimed water to Holmen Paper Company, which will become the first paper mill in Europe producing 100% recovered paper from 100% reclaimed water (4). In addition, plants located in Florence (Italy) and Villefranque (France) rely on MBR technology (179-180); whereas UF membranes were installed in Turin (Italy) and Katowice (Poland) (180-181).

All water reclamation projects that have been found within the African continent are located in *South Africa* (Figure 4), and all of them reclaim municipal wastewater for pulp and paper mills; although treatment characteristics are only available for Durban's water reclamation plant, which supplies 47,000 m^3/day of tertiary treated water (sedimentation + ozonation + activated carbon filtration + chlorination) to *Mondi* Paper Mill (182).

Finally, the USA gather most of the municipal wastewater reclamation projects aiming to supply process water to industrial applications that were found in America; whereas only two were found in Mexico, and another one in Canada. None were found located in Central and South America (Figure 4). Among them, there are two well differentiated types of facilities: a) those based on membrane technologies; and b) those consisting on tertiary treatments based on "*Californian Title 22*" recommendations or slight modifications of them. The main difference between both treatment schedules is whether the main objective is obtaining water just for cooling use, or if higher water quality standards are required, for which multi-barrier membranes systems would be installed (Table 6). In short, most plants located from Florida to Massachusetts produce make-up water for cooling operations through conventional tertiary treatments; whereas membrane filtration is preferred in those plants located from Middle to West USA (California and Arizona, mainly), and they are generally of bigger capacity. This may be expected provided California pioneered water reclamation initiatives creating Title 22 quality guidelines, and starting-up the first municipal facility using RO systems for water reclamation in 1977 (183).

5. GENERAL OVERVIEW AND DISCUSSION CONSIDERING ACTUAL FACILITIES RECLAIMING WATER WORLDWIDE

Conventional tertiary treatment (typically, flocculation + clarification + filtration + disinfection) is usually applied when reclaimed water would just be used for cooling applications; whereas membrane filtration is further required for the production of boiler make-up water at least, or when reclaimed water is going to be used within the process (Tables 3-6). Most of the facilities that use membrane systems to reclaim sewage rely on the combination of MF or UF with RO; and less are based on MBRs.

Although there were approximately 3,000 MBRs operating worldwide in 2008, the number of facilities that are nowadays applying this technology to reclaim municipal wastewater for industrial applications is still very low (Tables 3-6); although it is progressively increasing, and several pilot trials have been carried out to demonstrate the feasibility of combining MBR+RO technologies in new water reclamation projects (184-186).

Regarding NF treatment, it has been successfully used for reducing hardness, color and high organic content in feed water, at the same time that it represents an effective barrier for cysts and viruses (187). In comparison to RO, NF works at a lower operating pressure and at a higher permeate flux; but it only achieves partial nitrites/nitrates reduction (188), it reaches lower salts rejection results (e.g. 78% in NF versus 95% in RO), and COD would not totally be removed (189). Even considering RO treatment superiority, actual NF application to filtrate secondary effluents of municipal wastewaters is still very limited (190-191). Nevertheless, some good essays considering the implementation of NF technology to reclaim biologically treated wastewater from textile, and pulp and paper industries, have been reported to date (127, 192-194). In short, NF may be considered a promising water reclamation alternative to RO filtration in order to reduce the cost of the treatment in those applications where NF limitations would not be limiting the accomplishment of those defined target water quality requirements.

In general, and regardless the type of membrane being used, membrane processes are widely extended for treating industrial effluents and recycle them back into the process. For example, there are twenty Indian dyeing mills that have already implemented zero liquid discharge systems to recycle their effluents and reduce fresh water consumption; and even though each one has implemented its own specific wastewater treatment system, all of them use RO technology as the final treatment stage (156). Other companies within the textile industry, or other mills belonging to other industrial sectors, provide other good application alternatives. In short, it may be: an MBR treating bleaching water of cotton raw material (156); a combined UF+NF membrane system reducing freshwater use to its maximum extent in a mill producing fine and magazine quality papers (194-195); an MBR followed by an RO stage generating boiler feed water from a frozen-vegetables processing plant effluent (1); or an UF+RO membrane system implemented in order to satisfy boiler make-up water demand in a power plant (196). They all succeeded in recycling effluents inside their industrial processes keeping a competitive product fabrication cost.

Nevertheless, it is also very important to highlight one more the fact that using drinkable water quality in some industrial applications would not be necessary (e.g. cooling, or sealing applications). For example, some paper mills use clarified water from dissolved air flotation units for this purpose (197); and evaporator and boiler condensed streams can also be used as pump sealing water (198). In addition, other mills and facilities have reported the use of municipal tertiary treated effluent (biological oxidation + alum coagulation + filtration + disinfection) (199). In short, water and energy savings may turn crucial in the future against a higher water scarcity threshold that would be imposed under the foreseen climate change. On the other hand, even though it would not always be required to generate water of drinkable grade for many industrial applications, the inherent health hazard that is attributed to municipal sewage management will always be a major concern.

Industrial effluent recycling is probably the most extended option used to reduce freshwater consumption because the facilities are generally located far away from mWWTPs; and wastewater transport to a mill may not result economically feasible. Besides, municipal wastewater reclamation usually faces a general population disapproval, sometimes even for irrigation purposes (200), due to the presence of micro-pollutants (i.e. natural and synthetic hormones, pesticides, pharmaceuticals or personal care products), and microorganisms meaning potential health hazard. In short, final reclaimed water quality is always suspicious of carrying undesired hazards. As a consequence, many surveys have been performed in order to guess which would be the potential public willingness to use reclaimed sewage (201-202). Its main conclusion stated that the best way for promoting its acceptance is providing factual information regarding the production process, rather than launching persuasive campaigns. Moreover, in the same way people are nowadays concerned about plastic, glass, and paper recycling, the society should work on achieving the same concern for water use (industrial water use, particularly).

Summing up, Table 7 shows a summary of the general treatment removal efficiencies that are achieved for those main water quality parameters (e.g. TSS, TDS, COD, BOD_5, TOC, inorganic concentrations, biological contents, etc.) after applying different technological alternatives to reclaim municipal wastewater (4, 203). In short, Table 7 provide a first approximation to an appropriate reclamation treatment selection chart considering average municipal wastewater quality figures, and the requirements that should be considered for a specific industrial application, which were previously reviewed.

4. CONCLUSIONS

Environmental sustainability may be significantly enhanced reclaiming municipal wastewater. Particularly, it enables the replacement of fresh water use for industrial purposes in areas suffering water scarcity.

Depending on the type of industry, and on the specific application where reclaimed water is going to be used, water quality requirements that should be fulfilled are variable, so the implementation of those suitable reclamation technologies must be timely designed. The most important challenges that reclaimed facilities using membrane technologies must face are fouling and the management of the generated rejected streams.

The number of facilities that reclaim urban wastewater for different industrial applications has recently increased very quickly. Most of them rely on the use of multi-barrier membranes systems consisting of MF, UF or MBR units followed by RO systems, which are able to produce water of a very high quality (drinkable grade).

Screening worldwide mWWTPs relaying on the use of membrane systems for producing reclaimed water for industrial applications, it has been found that 19 membrane filtration-based facilities over a total 28 water reclamation plants are currently located in Asia (6 of them placed in Singapore); 15 of 45 in America, 9 of 10 in Australia, and 4 of 6 in Europe. Only four plants, all of them related to paper mills, were found reported in South Africa, but it could not be verified whether they apply membrane technologies. No sewage reclamation facilities were currently found located in Central and South America.

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Table 1. Quality guidelines that reclaimed water must fulfill for its use as fresh water in different industrial applications.

				PAPER GRADES (61)						**TEXTILE (5)**					
Parameter	**Cooling (37)**	**Boiler (37, 38)**	**Sealing (55)**	**Groundwood**	**Soda & sulphate (Kraft)**	**Kraft bleached**	**Unbleached**	**Bleached**	**Recycled (high pressure showers)**	**Sizing suspension**	**Scouring, bleaching, dying**	**Electronic (83)**	**Chemical (88)**	**Pharmaceutical [a] (56)**	**Petrochem / Coal (88)**
T,ºC	-	-	<20-30	<55	27	-	-	15-27		-	-	19-21	-	-	-
pH	6.9-9.0	8.2-10.0	>7.0	-	-	-	-	-	6.5-7.5	-	-	6.5-7.5	6.2-8.3	-	6-9
TSS, mg/L	100	5	-	40	10	10	10-30	10-30	<5	5	5	-	5	-	10
TDS, mg/L	500	700	<1000	250-1000	250-1000	300	75-650	75-650	<300	100	100	-	1000	-	1000
Conductivity, mS/cm	-	-	<2.0	-	-	-	-	-	<0.5	-	-	-	-	≤1.1	-
Turbidity, NTU	50	-	-	70	35	40	140	14-56	-	-	-	-	-	-	-
Colour, PCU	-	-	-	30	5	25	30-100	5-25	<30	5	5	-	20	-	-
COD, mg/L	75	5	-	-	-	-	-	-	<5	-	-	-	-	-	-
BOD_5, mg/L	25	-	-	-	-	-	-	-	-	-	-	-	-	-	-
TOC, mg/L	-	-	-	-	-	-	-	-	-	-	-	0.25	-	0.5	-
Hardness, $mgCaCO_3/L$	650	1.0	<200	100-200	100-200	100	200	100	<200	25	25	-	250	-	350
Alkalinity, $mgCaCO_3/L$	350	100	-	75-150	75-150	75	150	75-125	<100	-	-	-	125	-	-
Ammonia-N, mg/L	1.0	0.1	-	-	-	-	-	-	<0.5	-	-	-	-	-	-
PO_4^{3-}, mg/L	4.0	-	-	-	-	-	-	-	-	-	-	-	-	-	-
HCO_3^- , mg/L	24	120	-	-	-	-	-	-	-	-	-	-	128	-	-
NO_3^-, mg/L	-	-	-	-	-	-	-	-	-	-	-	-	5	≤0.2	-
Si, $mgSiO_2/L$	50	10	-	50	20	50	100	9-20	<5	-	-		50	-	-

[a]Data are for highly purified water class; [b]Heavy metals in general must be ≤ 0.1 mg/L.

Table 1 (continuation). Quality guidelines that reclaimed water must fulfill for its use as fresh water in different industrial applications.

				PAPER GRADES (61)						TEXTILE (5)					
Parameter	**Cooling (37)**	**Boiler (37, 38)**	**Sealing (55)**	**Groundwood**	**Soda & sulphate (Kraft)**	**Kraft bleached**	**Unbleached**	**Bleached**	**Recycled (high pressure showers)**	**Sizing suspension**	**Scouring, bleaching, dying**	**Electronic (83)**	**Chemical (88)**	**Pharmaceutical [a] (56)**	**Petrochem / Coal (88)**
Al, mg/L	0.1	0.1	-	-	-	-	-	-	0.1	-	-	-	-	-	-
Fe, mg/L	0.5	0.3	-	0.3	0.1	0.2	1.0	0.1	<0.1	0.3	0.1	-	[b]	0.1	1.0
Mn, mg/L	0.5	0.1	-	0.1	0.05	0.1	0.5	0.03	<0.05	0.05	0.01	-	[b]	0.1	-
Ca, mg/L	50	0.4	-	-	-	-	-	-	<60	-	-	-	-	68	75
Mg, mg/L	0.5	0.25	-	-	-	-	-	-	<15	-	-	-	-	19	30
SO_4^{2-}, mg/L	200	-	<200	trace	-	-	-	100-300	<100	-	-	-	-	100	-
Cl^-, mg/L	500	-	<200	75	75	200	200	200	<50	-	-	-	-	500	300
Cu, mg/L	-	0.05	-	-	-	-	-	-	<0.001	0.01	-	-	[b]	-	0.05
Zn, mg/L	-	0.01	-	-	-	-	-	-	-	-	-	-	[b]	-	-
Na, mg/L	-	-	-	-	-	-	-	-	-	-	-	0.01	-	-	-
Dissolved oxygen, mg/L	-	0.007	-	-	-	-	-	-	-	-	-	-	-	-	-
Methylene-blue active substances	-	1	-	-	-	-	-	-	-	-	-	-	-	-	-
Carbon tetrachloride extract	-	1	-	-	-	-	-	-	-	-	-	-	-	-	-
Evaporation residue, mg/L	-	-	-	-	-	-	-	-	-	-	-	0.5	-	-	-
Resistivity, MΩ·cm at 25ºC	-	-	-	-	-	-	-	-	-	-	-	>18	-	-	-
Abrassive matter (>10µm)	-	-	<40-50	-	-	-	-	-	-	-	-	-	-	-	-
Aerobic bacteria (CFU/mL)	-	-	-	-	-	-	-	-	-	-	-	-	≤10	-	-
Bacteria endotoxins (IU/mL)	-	-	-	-	-	-	-	-	-	-	-	-	≤0.25	-	-

[a] Data are for highly purified water class; [b] Heavy metals in general must be ≤ 0.1 mg/L.

Table 2. Effect of common foulants on membrane performance (104, 106-109).

FOULANT TYPE	SALT PASSAGE	PRESSURE DROP	PRODUCT FLOW
Inorganic salts	10-15% increase	10-40% increase	<10% decrease
Metal (hydro)oxides	>200% rapid increase	>200% rapid increase	20-40% decrease
Colloids	>200% gradual increase	>200% gradual increase	>50% gradual increase
Organic matter	increase or decrease	small increase	>50% decrease
Biofouling	> 200% rapid decrease	>200% rapid increase	>50% decrease

Table 3. Wastewater reclamation facilities based on membrane systems located in Australia.

FACILITY NAME	LOCATION	SYSTEM	APPLICATION
Bundamba AWTP[a] (162)	Ipswich	MF + RO + UV	Cooling water
Eraring Power Station (1)	Lake Macquarie	MF + RO[e]	Boiler makeup water Cooling water
Gibson Island AWTP[a] (163)	Brisbane	MF + RO + UV	Power stations
Kwinana Water Reuse Project (204)	Perth	MF + RO[e]	-[d]
Luggage Point WTP[b] (160)	Brisbane	MF + RO[e]	-[d]
Luggage Point AWTP[a] (161)	Brisbane	MF + RO[e]	Boiler feed water Cooling tower make-up Other process uses
Northern Water Plant (205)	Victoria	UF + RO[e]	Refinery
Illawarra RWP[c] (160)	Sydney	MF + RO[e]	At Bluescope Steel facility
WRAMS-2000 Olympic Site (206)	Sydney	MF + RO + UV	-[d]

Secondary treatment is specified when RO is used without membrane pretreatment. [a]AWTP: Advanced Water Treatment Plant; [b]WTP: Water Treatment Plant; [c]RWP: Recycled Water Plant; [d]Industrial, but not specified; [e]Not specified whether disinfection exists after RO.

Table 4. Wastewater reclamation facilities based on membrane systems located in Asia.

FACILITY NAME	**LOCATION**	**SYSTEM**	**APPLICATION**
Bedok NEWater plant (23, 207)	Bedok/Singapore	UF + RO + UV	Electronics industry Wafer fabrication Air conditioning Cooling water Boiler feed water
Kranji NEWater plant (23, 209)	Kranji/Singapore	MF + RO + UV	Electronics industry Wafer fabrication Air conditioning Cooling water Boiler feed water
Seletar NEWater plant (23, 209)	Seletar/Singapore	MF + RO + UV	Electronics industry Wafer fabrication Air conditioning Cooling water Boiler feed water
Changi NEWater (210)	Changi/Singapore	UF + RO + UV	Electronics industry Wafer fabrication Air conditioning Cooling water Boiler feed water
Ulu Pandan NEWater plant (23, 186)	Ulu Pandan/Singapore	MF + RO + UV	Electronics industry Wafer fabrication Air conditioning Cooling water Boiler feed water
Xinxinban WWTP[a] (209)	Hohhot/China	MBR + IE[l]	Cooling water

Secondary treatment is specified when RO is used without membrane pretreatment.[a]WWTP: Wastewater Treatment Plant; [b]WRP: Water Recycling Plant; [c]STP: Sewage Treatment Plant; [d]Industrial, but not specified; [e]Not specified whether disinfection exists after the last treatment; [f]SF: Sand filter; [g]EDI: Electrodeionization; [h]CMF: Classical matched filter; [i]CF: Cartridge filter; [j]GF: Gravity filter; [k]AC: Activated carbon; [l]IE: Ionic exchange.

Table 4 (continuation). Wastewater reclamation facilities based on membrane systems located in Asia.

FACILITY NAME	LOCATION	SYSTEM	APPLICATION
Jurong WRP[b] (210)	Jurong Island/Singapore	SF[f] + RO[e]	-[d]
Jurong WRP[b] (210)	Jurong Island/Singapore	MBR[e]	-[d]
CNPC Dalian Petrochemical Company Ltd. (211)	Dalian/China	UF + RO[e]	Boiler feed water
ShanXi DaTong Co-gen Ltd. (212)	DaTong/China	UF + RO + EDI[g]	Boiler feed water
Tianjin Ji Zhuang Zi STP[c] (8)	Tianjin/China	MF[e]	-[d]
Tianjin TEDA STP[c] (8)	Tianjin/China	MF + RO[e]	-[d]
STP of Beijing EDA (176)	Beijing/China	CMF[h] + RO[e]	Process water
Gajwa WWTP (9)	West Inchen/North Korea	Flocculation + media filtration + RO[e] (MF is applied to protect RO against process breakdown)	Cooling water
Sewage Treatment of Changshou industry park (176)	Changshou/China	UF[e]	Cooling water Boiler make-up water Industrial process water
Madras Fertilizers Limited (8)	Chennai/India	SF[f] + ammonia stripping + carbonation + chlorination + filtration +dechlorination +CF[i] +RO[e]	Cooling tower makeup
Petromin Refinery (Aramco) (213, 214)	Riyadh/Saudi Arabia	Lime clarification + GF[j] + AC[k] + RO + decarbonation + IE[l]	Cooling tower makeup Desalter water makeup Boiler feed water
Sulaibiya WWTP (215, 216)	Sulaibiya/Kuwait	UF+RO+UV	-[d]

Secondary treatment is specified when RO is used without membrane pretreatment. [a]WWTP: Wastewater Treatment Plant; [b]WRP: Water Recycling Plant; [c]STP: Sewage Treatment Plant; [d]Industrial, but not specified; [e]Not specified whether disinfection exists after the last treatment; [f]SF: Sand filter; [g]EDI: Electrodeionization; [h]CMF: Classical matched filter; [i]CF: Cartridge filter; [j]GF: Gravity filter; [k]AC: Activated carbon; [l]IE: Ionic exchange.

Table 5. Wastewater reclamation facilities based on membrane filtration systems located in Europe.

FACILITY NAME	**LOCATION**	**SYSTEM**	**APPLICATION**
Cuenca Media-Alta de Arroyo Culebro WWTP[a] (4)	Madrid/Spain	SF + AC + UF + RO + UV	Process water in a paper mill Cooling tower makeup Boiler feed water
Flag Fen Sewerage Treatment Plant (217)	Peterborough/UK	MF+ chloramines +RO +IE	Boiler feed water
E.C. Katowice Power Plant (181)	Katowice/Poland	UF	Cooling water
Villefranque (180)	Villefranque/France	MBR	Washing water in a tannery
Empoli MWRF (179)	Florence/Italy	MBR	Industrial (but not specified)
Collegno (180)	Turin/Italy	SF + UF + AC	Industrial (but not specified)

Secondary treatment is specified when RO is used without membrane pretreatment [a]WWTP: Wastewater Treatment Plant; [b] *Type of disinfection not specified; ** Not specified whether disinfection exists after RO. MWRF: Municipal Water Reclamation Facility. SF: Sand filtration; CF: Cartridge filtrate; IE: Ionic exchange; AC: Activated carbon; DMF: Dual media filters.

Table 6. Wastewater reclamation facilities based on membrane systems located in America.

FACILITY NAME	LOCATION	SYSTEM	APPLICATION
South Bay ARWTF[a] ICF (218)	Sta. Clara & S. Jose/ California	MF+RO+UV	Cooling and processing
Toppan Electronic WWTP[b] (219)	San Diego/California	MF + RO[d]	Process water
Millender-McDonald Carson Regional WRP[c] (8)	Carson/California	MF + RO[d]	Cooling tower makeup
Mobil Boiler Feed Facility (8)	Torrance/ California	MF + RO[d]	Boiler feed water
West Basin WRP[c] (220)	El Segundo/California	MF + RO[d]	Boiler feed water
Terminal Island Treatment Plant (8)	Los Angeles/California	MF + RO + chlorination	Boiler feed water
Pinellas County Resource Recovery Facility (221)	Pinellas/Florida	MF + RO + chlorination	Boiler feed water
City of North Las Vegas WRP[c] (222)	Las Vegas/Nevada	MBR + chlorination	Cooling water
Kyrene WRP[c] (223)	Tempe/Arizona	MBR + UV	Industrial processing
Wyodak Power Plant (200)	Gilette/Wyomin	SF +CF +RO + recarbonation +IE	Boiler make-up Dust suppression
Redbud Power Plant (224)	Luther/Oklahoma	Secondary + RO[d]	Cooling tower makeup Boiler feed water
Harlingen WWTP[b] (225)	Harlingen/Texas	SF + RO[d]	Process water
Honouliulu WWTP[b] (226)	Eva Beach/Hawaii	MF + RO + disinfection[e]	Boiler feed water
City of Edmonton Gold Bar WWTP[b] (227)	Edmonton/Canada	UF + RO[d]	Hydrogen and steam production Process alternate feedstocks
Met-Mex Peñoles (228)	Torreon/Mexico	DMF[f] + AC[g] + CF[h] + RO[d]	Cooling tower makeup Boiler feed water Zinc electrolytic process

Secondary treatment is specified when RO is used without membrane pretreatment. [a]ARWTF: Advanced Recycled Water Treatment Facility; [b]WWTP: Wastewater Treatment Plant; [c]WRP: Water Reclamation Facility; [d]Not specified whether disinfection exists after RO; [e]Type of disinfection not specified; [f]DMF: Dual media filters; [g]AC: Activated carbon; [h]CF: Cartridge filter.

Table 7. Removal efficiencies (%) of different treatments applied to reclaim municipal sewage (4, 203).

Parameter	**CAS**[a]	**CAS + filtration**	**CAS +BNR**[b]	**CAS +BNR + filtration**	**MBR**	**MBR +IE**	**CAS +MF / UF +RO MBR+RO**
TSS, mg/L	96-94	98	95-96	99	>98	>98	>99
TDS, mg/L	0	0-19	0-19	0-19	0-19	-	85-98
VOCs, µm	90	90	90-95	90-95	90-95	90-95	>99
COD, mg/L	84-90	88-91	92-95	92-96	>96	>96	96-99
BOD_5, mg/L	93-95	94-95	95-96	98-99	>99	>99	>99
TOC, mg/L	85-88	88-90	90-92	98-99	>98	>98	99.0-99.9
Total nitrogen, mg/L	25-50	25-50	85-89	90-93	>86[c]	>80	>95
Total phosphorous, mg/L	0-17	0-33	75-83	>83	58-93[d]	>80	>86
Metals, mg/L	33-40	33-40	33-40	33-40	trace	trace	-
Total coliforms, CFU/100mL	99.0-99.9	>99.9	99.0-99.9	99.0-99.9	>99.9	>99.9	~100
Protozoan cysts and oocysts, CFU/100 mL	0-99.9	>99.9	>99.9	>99.9	>99.9	>99.9	~100
Viruses, PFU/100mL	0-90.0	0-99.9	0-90.0	0-90.0	>90	>90	~100

[a]CAS: conventional activated sludge + nitrification. [b]BNR: biological nutrient (N and P) removal ; [c]With anoxic stage; [d]With coagulant addition.

Figure 1. Water consumption for cooling towers at thermoelectric power plants (38).

Figure 2. Maximum and minimum fresh water consumption values in paper mills (55) and textile industry (65). Data are scaled by tonne of final product.

Figure 3. Water consumption thresholds (maximum and minimum) within food industry (74). Data are scaled per tonne of product*, or per tonne of raw material**.

Figure 4. Worldwide distribution of municipal water reclamation facilities where reclaimed water is used for industrial activities.

Figure 1

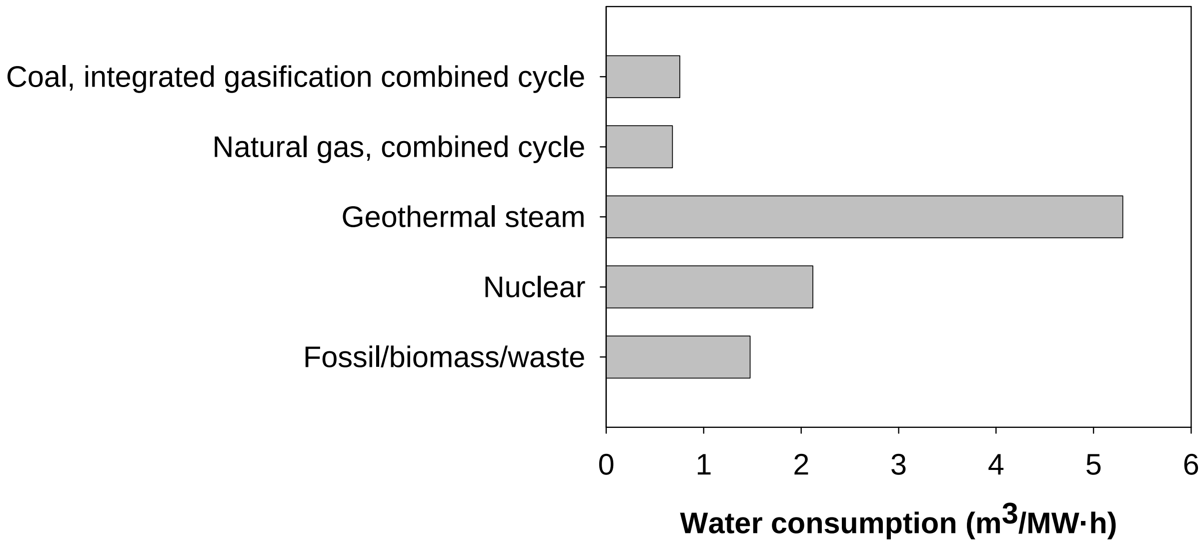

Coal, integrated gasification combined cycle
Natural gas, combined cycle
Geothermal steam
Nuclear
Fossil/biomass/waste
0
1
2
3
4
5
6
Water consumption (m3/MW·h)

Figure 2

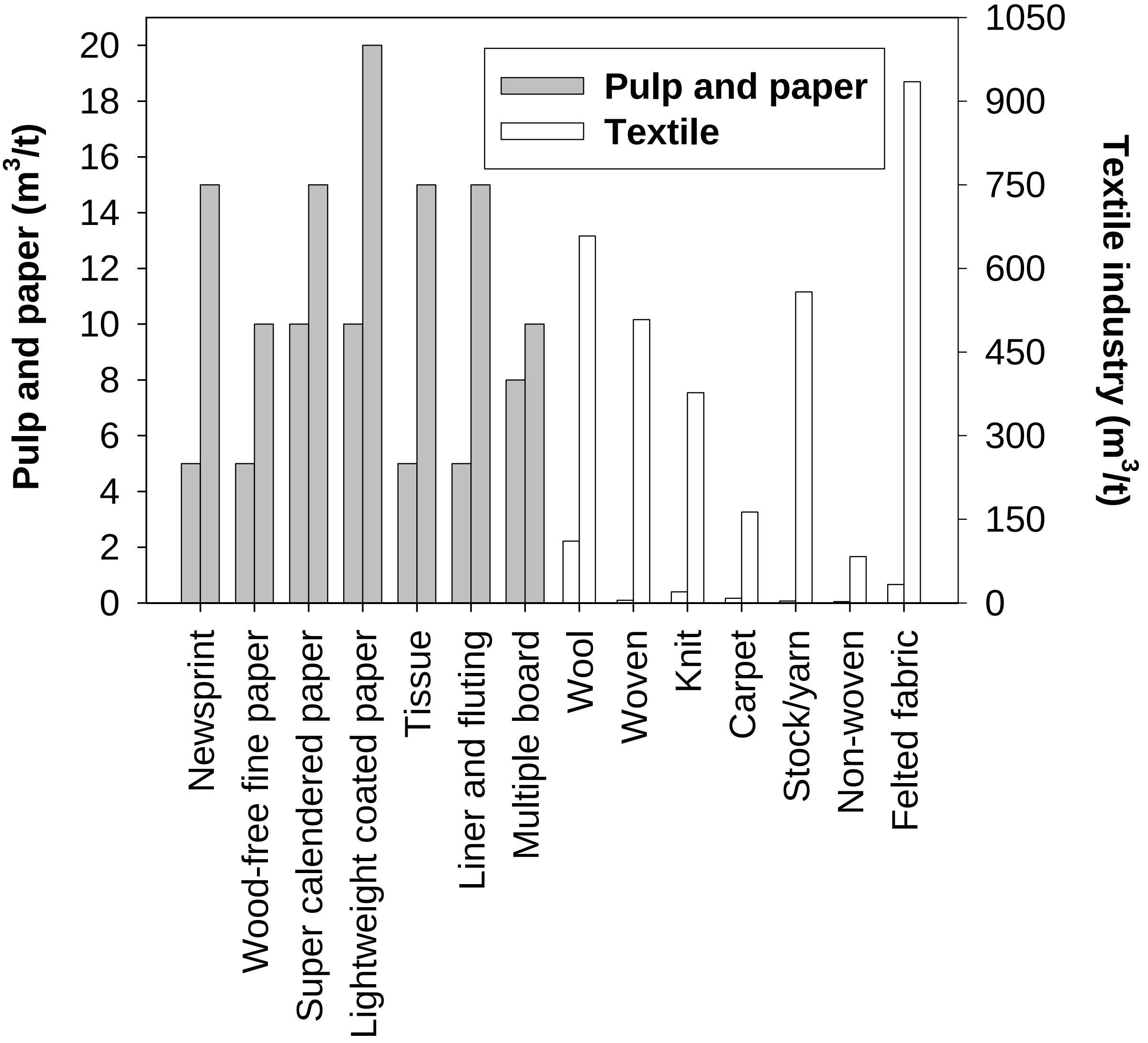

Pulp and paper
Textile
Pulp and paper (m³/t)
Textile industry (m³/t)
0
2
4
6
8
10
12
14
16
18
20
0
150
300
450
600
750
900
1050
Newsprint
Wood-free fine paper
Super calendered paper
Lightweight coated paper
Tissue
Liner and fluting
Multiple board
Wool
Woven
Knit
Carpet
Stock/yarn
Non-woven
Felted fabric

Figure 3

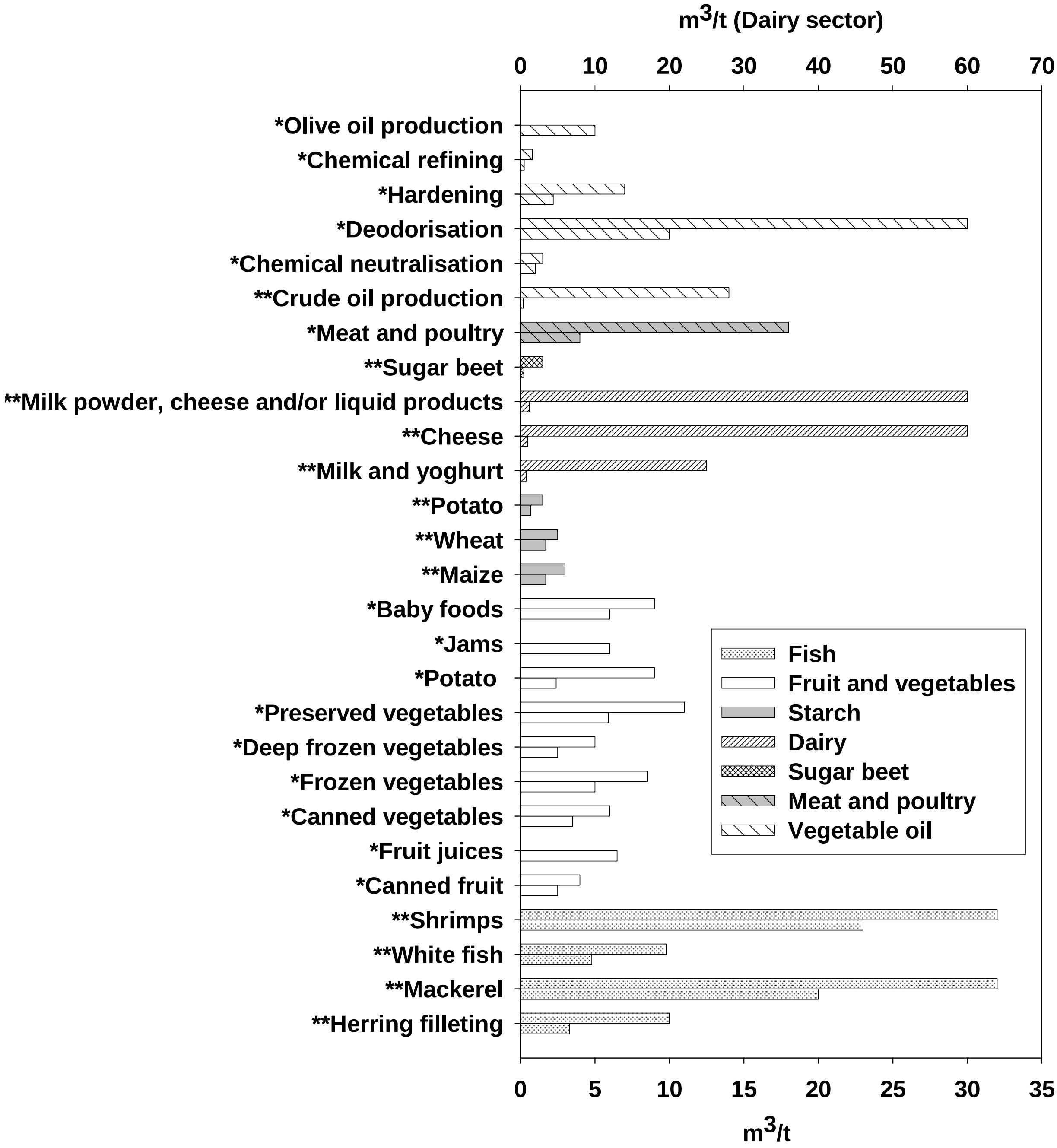
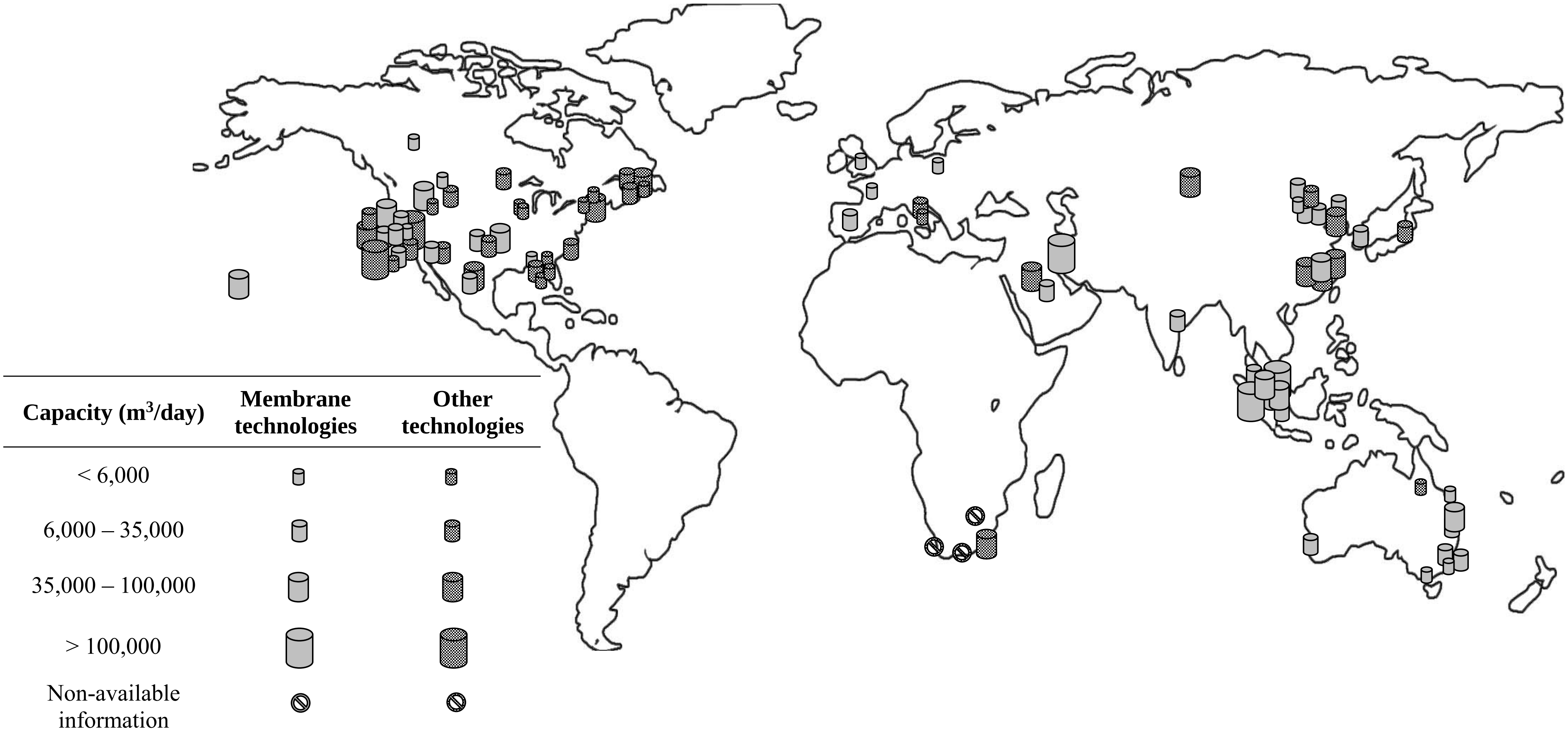

m3/t (Dairy sector)
0
10
20
30
40
50
60
70
*Olive oil production
*Chemical refining
*Hardening
*Deodorisation
*Chemical neutralisation
**Crude oil production
*Meat and poultry
**Sugar beet
**Milk powder, cheese and/or liquid products
**Cheese
**Milk and yoghurt
**Potato
**Wheat
**Maize
*Baby foods
*Jams
*Potato
*Preserved vegetables
*Deep frozen vegetables
*Frozen vegetables
*Canned vegetables
*Fruit juices
*Canned fruit
**Shrimps
**White fish
**Mackerel
**Herring filleting
Fish
Fruit and vegetables
Starch
Dairy
Sugar beet
Meat and poultry
Vegetable oil
0
5
10
15
20
25
30
35
m3/t

Figure 4


Capacity (m³/day)
Membrane technologies
Other technologies
< 6,000
6,000 – 35,000
35,000 – 100,000
> 100,000
Non-available information