Priority pollutants removal in an MBR-RO wastewater treatment system

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Abstract

A small footprint wastewater treatment plant that consists of a membrane bioreactor coupled with a reverse osmosis unit (MBR-RO) has been placed and set in operation for 12 months in EYDAP's R&D department in order to evaluate the quality of the effluent and to explore the feasibility of reuse of the reclaimed water as specified in the Greek National legal framework. A sustainable technology called sewer mining has been approached, which abstracts raw wastewater directly from the sewerage network, treats it on site and provides water at the point of demand. Monitoring of system's performance was achieved through a series of lab analyses and on-line measurements. Besides the microbiological and conventional parameters, final effluent was also analyzed for heavy metals, priority pollutants and emerging contaminants in order to examine compliance with the threshold values set in Greece in order to allow for wastewater reuse for Wastewater Treatment Plants with a population equivalent greater than 100,000. Results showed that the MBR-RO technology provides for the achievement of a high quality effluent, suitable for many reuse purposes. The research confirmed the need for RO as a post treatment level in the case of saline wastewater and/or very strict threshold values for organic micropollutants.

Keywords: Sewer mining, Membrane bioreactor, Reverse osmosis, Emerging contaminants, Priority pollutants, heavy metals

1. Introduction

The single most important factor that determines the success of a water treatment process is the effluent wastewater's quality. There are numerous quantifiable parameters that collectively assess quality, some of which are indirect and act as indices of the presence of biochemical substances, while others are of direct nature, measuring a specific compound or microorganism. From the latter, priority pollutants (PPs) and heavy metals are of paramount importance in wastewater urban reuse, where the legal framework regarding urban reuse is gradually becoming stricter worldwide. PPs are substances that pose a danger to both the environment and human health and may be present in water (Kislenko, et al., 2011). Within these substances several groups of compounds can be identified such as Organotins, Volatile Organic Chemicals (VOCs), Polycyclic Aromatic Hydrocarbons (PAHs), Alkylophenols, Pesticides, Chlorobenzenes, Phthalates and others (Gasperi, et al., 2008). On the other hand, the most commonly detected toxic heavy metals in wastewater include Arsenic (As), Lead (Pb), Mercury (Hg) and Cadmium (Cd), Copper (Cu), Chromium (Cr), Nickel (Ni),Silver (Ag) and Zinc (Zn) (Akpor, et al., 2014). These metals pose a serious threat to humans and to the aquatic environment since they can be absorbed, accumulated and biomagnified and can cause several known diseases, due to their toxic nature above certain thresholds (Herojeet, et al., 2015). Moreover it has been found that they can affect several organs such as the kidney and induce malfunctions to the neurological system (Lohami, et al., 2008). It has to be noted that Cd, Hg and Pb are highly toxic to humans and animals but are less toxic to plants, while Zn, Ni and Cu are, when present in excess concentrations, more damaging to plants than to humans and animals (Tiruneh, et al., 2014). Heavy metals and PPs can enter a municipal sewage network through various paths such as water runoff, groundwater and sanitary, light industrial, domestic or commercial sewage. Several past researches have investigated the source of both PPs (Soonthornnonda & Christensen, 2008); (Rule, et al., 2006), and heavy metals (Sorne & Lagerkvist, 2002). The main legislative tool that is being used for the protection of the aquatic environment as well as for armoring water quality, within the European Union (EU), is the Water Framework Directive (2000/60/EC) (WFD). Article 16 of the WFD develops the European Union (EU) strategy against pollution of water by chemical substances. A list of 33 priority substances has been established and most of the list's entries are organic contaminants (hydrocarbons, organochlorine compounds, organic solvents, pesticides, and chlorophenols), four of them are toxic metals and one is an organometallic compound (tributyltin). Moreover, WFD makes a distinction between priority substances, for which their emissions should be reduced as far as possible and priority hazardous substances, the use of which should be ceased or emissions, discharges and losses should be phased out by 2020. Priority hazardous substances are toxic, persistent

and have the tendency to bio-accumulate. However it has to be mentioned that there is no certain definition for determining priority substances (Kislenko, et al., 2011). The first list in the WFD was replaced by Annex II of the Directive on Environmental Quality Standards (Directive 2008/105/EC), which set environmental quality standards (EQS), while Directive 2009/90/EC layed down technical specifications for chemical analysis and monitoring of water status and introduced a list of 11 substances under review for being future entrances in the PP list. Last of the directives was Directive 2013/39/EU, which brought further additions to the former and updated the initial list of 33 PPs, adding another 12 elements, compiling a list of a total of 45 compounds. This continious upgrade of the EU directives highlights the importance of the water quality standards applied and gives insight to future directions. The Greek legislation regarding wastewater reuse introduces certain quantitative limits which depend on the end use of the water. More specifically, the limit values specified in the Greek National legislation regarding wastewater reuse for unrestricted irrigation and urban use were introduced by the JMD 145116/2011. Regarding heavy metals and PPs, there are two separate tables for each category which incorporate 19 and 40 compounds respectively. An amendment of the Joint Ministerial Decision 145116/2011 (JMD) occurred via the Government Gazette B 69/2016 (GG), which introduced 3 new PPs and more detailed quality standards. In this context, monitoring of heavy metals and PPs becomes crucial in applications of water reclamation. One of the recycling technologies that steadily gains popularity among those technologies aiming to substitute fresh water in non-potable uses is called sewer mining (SM). This practice focuses on draining wastewater directly from the sewage network, while the treatment takes place at the point of use. It belongs to the group of decentralized options for water recycle/reuse, targets mainly urban reuse and, therefore, it is considered appropriate to examine an even wider spectrum of substances, including emerging contaminants (Sauve & Desrosiers, 2014) such as various compounds that belong to the endocrine disrupting chemicals (EDCs) and the non-steroidal anti-inflammatory drugs (NSAIDs), which, despite not being part of the list of PPs yet, have gained a lot of attention in research due to their persistent detection in the aquatic environment and their possible adverse effects. For the goal of examining the quality of the reclaimed wastewater of a SM unit, an innovative small footprint SM packaged treatment unit for urban reuse, consisting of a membrane bioreactor (MBR) coupled with a reverse osmosis (RO) unit, has been installed in EYDAP's R&D department, in the Metamorphosis region (Athens, Greece). Athens demo site tests the idea of SM as a concept for distributed reuse within the urban environment, exploiting state-of-the-art information and communication technology solutions for distributed monitoring and management. In view of the above, the objective of this study was to examine the presence of certain heavy metals, PPs and emerging contaminants in municipal wastewater and to access the performance of an MBR-RO pilot system in relation to the efficiency of their removal.

2. Materials and methods

2.1. Description of the MBR-RO pilot system

Dual-membrane processes, such as an ultrafiltration (UF) with an RO, are gaining popularity in the process of reclaiming municipal wastewater, due to their efficiency and simplicity in operation. The role of the UF membranes is to perform secondary treatment of wastewater and RO acts as the polishing treatment step. The suspended solids are removed by UF membranes while RO membranes remove dissolved solids, organic and ionic matter. A membrane bioreactor (MBR) can carry out the secondary treatment of sewage and produces an effluent that fulfills the qualitative criteria for being fed to an RO unit, and hence MBR-RO has a great potential for the treatment of raw sewage to produce reclaimable water (Comerton, et al., 2005); (Xiao, et al., 2014). The schematic presentation of the unit is provided in Figure 1. Feed wastewater is pumped from the local sewerage network to the satellite wastewater treatment plant (WWTP). The inlet pumping station is directing the sewage, via a preliminary treatment that includes a compact fine screen-grit system and a biotube filter, in the equalization tank of the system. From there, the degritted sewage is overflowing to the main treatment units. The main treatment units consist of biological treatment with MBR and finally an RO unit. The denitrification stage is the first to take place inside an anoxic tank equipped with a proper mixing device. The mixed liquor from the denitrification tank enters the aeration tank, where the biological processes of oxidation of the organic load, nitrification and stabilization of sludge occur. Separation of the suspended solids from the treated effluent is taking place through an ultrafiltration membrane. The installed membrane consists of ultrafiltration modules that operate under negative pressure, with a filtration direction going from the outside towards the inside of the modules. Solids are therefore withheld in the retentate on the outside, while the permeate flows inside and is directed by a lobed pump to a permeate accumulation tank, while excess sludge returns to sewage network. From that accumulation tank, the permeate ends up to the RO system. The need for RO as a post treatment level derives from the necessity to comply with the environmental standards as in the case of saline wastewater. Moreover, the unit has the ability to work. without RO treatment, in which case the permeate ends up directly into the effluent tank. Table 1 sums up the characteristics of both the MBR and RO membranes as well as those of the pilot unit. Cleaning of the membranes with air is performed through an aeration system that consists of blowers and coarse bubble diffusers, thus protecting the membranes from fouling and particle deposition. Moreover, in order to maintain membrane permeability, two more methods of membrane cleaning have been applied. The first one is the backflushing mode, where the lobbed pump inverts its rotation sense and conveys a part of the produced permeate from the inside to the outside of the UF modules so as to detach excess material. The second one is maintenance cleaning; chemical cleaning cycles consisting of sodium hypochloride (NaOCl) and citric acid, that reach the

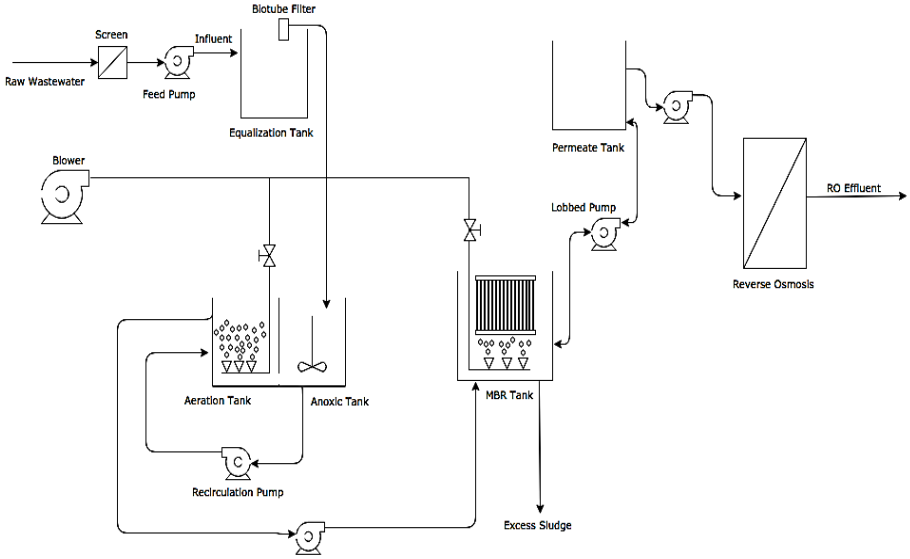

Figure 1. Schematic presentation of the MBR-RO unit

Table 1. Membrane and pilot system characteristics

Membrane characteristics	**MBR**	**RO**	**Pilot parameters**	**MBR**	**RO**
Manufacturer	KOCH Membrane systems	Filmtech membranes	**Manufacturer**	Chemitec	Chemitec
Module type	PSH 34	XLE 4040	**Configuration**	Hollow fiber	Spiral wound
Nominal pore size	0.03μm	-	**Operation mode**	Continuous	
Surface area	$34m^2$	$8.1m^2$	**Permeate volume(m^3 d^{-1})**	10	-
Material	PVDF	Polyamide Thin-Film Composite	**Coarse bubble aeration rate(m3 h-1)**	18	-
Salt rejection	-	99%	**Operating pressure(bar)**	-0,6 to 0.6	3 to 10

membranes by backflushing clean water that is enriched with those chemicals through dosage pumps.

2.2. Analytical methods

Wastewater characteristics (chemical oxygen demand, biochemical oxygen demand, total suspended solids, total volatile solids, sludge volume index, total phosphorus, total nitrogen, ammoniacal and nitrate nitrogen, chlorides, total and fecal coliforms and *E. coli*) were determined according to Standard Methods (American Puplic Health Association, 2012). For the determination of the selected emerging contaminants, wastewater samples were analyzed using a chromatographic method developed by Samaras *et al*. (2011). The developed procedure included solid phase extraction, while for the qualitative and quantitative analysis, an Agilent Gas Chromatograph 7890A connected to an Agilent 5975C Mass Selective Detector (MSD) was used. Furthermore, for the detection of heavy metals in the inlet flow and the MBR effluent, the method that was used was inductively coupled plasma mass spectrometry (ICP-MS), while for the RO effluent the selected method was Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES). For the PPs four different approaches were followed, depending on the chemical; Purge and Trap Gas Chromatography–Mass Spectrometry (T&P/GC-MS), Gas Chromatography coupled to tandem Mass Spectrometry (GC-MS/MS), Liquid Chromatography coupled to tandem Mass Spectrometry (LC-MS/MS).

3. Results and discussion

3.1. Untreated wastewater characterization

The influent of the pilot unit was tested for the occurrence of certain emerging contaminants belonging to the EDCs and NSAIDs (Figure 2), as well as for all heavy metals, trace elements and PPs that are specified in the Greek

legislation for water reuse. In the first group of chemicals, all components were found to be present in the influent stream, with concentrations ranging from 0.2 μg L^{-1} up to 8.8 μg L^{-1}. From those, the only pollutant that is subject to legislation is Nonylphenol 4 (NP), which happens to be the most abundant in the examined wastewater. According to a previous study, NP concentration is higher in wastewater coming from runoff samples near light industries (Rule, et al., 2006). NP has been found to cause inhibition of wheat growth, affect chlorophyll and several enzymes and thus is highly toxic for wheat and probably for many other plants (Zhang, et al., 2016). Therefore, NP should be monitored regularly, especially in winter months where it has been found that influent concentrations are higher (Gao, et al., 2017). Concerning heavy metals and trace elements, the chemicals under investigation were the ones specified in JMD 145116/2011 with one addition; silver (Ag). From those 20 compounds, only nine were identified in the examined wastewater, while all the other analyzed compounds were found to range in values below their respective limit of detection of the analytical method (LOD), which was 0.005 μg L^{-1} or 0.001 μg L^{-1}, depending on the element. The detected metals were Aluminum (Al), Copper (Cu), Iron (Fe), Lithium (Li), Manganese (Mn), Nickel (Ni), Lead (Pb), Vanadium (V) and Zinc (Zn). In a study performed by Sorne & Lagerkvist (2002), regarding urban wastewater, it was found that Cu mostly derives from households, specifically from copper pipes and taps. In the case of Zn, the load is equally divided between households and businesses (mainly car wash enterprises), while Pb originates mostly from commercial activities. Another study suggests that in the case of Cu, Pb and Zn, light industrial sources own a greater diffusion share (Rule, et al., 2006). Moreover, the influent analysis comes in good agreement with other studies regarding the ranking of concentration magnitude of metals in raw wastewater. More specifically, the occurrence of heavy metals in urban wastewater seems to follow –with slight variations- this sequence: Fe>> Al> Zn> Mn> Cr> Cu> Ni> Pb> Cd (Ustin, 2009); (Gulyas, et al., 2015); (Karvelas, et al., 2003). The influent metal concentration order produced from this study is Fe> Al> Zn> Mn> Cu> Pb> Ni> Li> V, so the only obvious difference is that Pb has a greater concentration in the studied sample, while Cr was below its LOD. Regarding PPs, from a total of 45 compounds, only four were present in the wastewater sampled. More specifically, these were Chloroform ($CHCl_3$), Trichloroethylene (TCE), Tetrachloroethylene (PCE) and Benzene (C_6H_6), none of which is considered as a hazardous priority substance. All of the aforementioned compounds belong to the VOC PPs. Concerning $CHCl_3$, a study of Rule *et al* (2005), has found that it is the only solvent that was found to have concentration greater than its LOD on domestic level. While Chloroform concentration in that study was found greater in domestic sewage, for TCE and PCE the authors suggest that dry cleaning was the reason why their concentration was greater in samples retrieved from the town center, where commercial activities take place. For $CHCl_3$, another study indicates that it has a far greater concentration in the water supply in comparison to domestic sewage, proposing that chlorination must be the main source of chloroform in wastewater (Wilkie, et al., 1996). Finally, other studies suggest that the four compounds that were found in the wastewater sample are usually undetected or found around the quantification limit (Gasperi, et al., 2009).

3.2. MBR-RO removal efficiency

The operational parameters of the unit at the time of the sampling are presented in Table 2. Moreover, results concerning nutrients, organic and microbial load are demonstrated in Table 3. It is evident that the system managed to complete eliminate the microbial load and achieve high removal of organic and inorganic contaminants Concerning the presence of the selected contaminants, Figure 2 illustrates the concentration of all 8 EDCs and NSAIDs, while Figure 3 presents the contribution to removal of the RO and the MBR units respectively.It is clear that the system manages to completely remove all substances except from the Nonyphenols, which, however, have a value of less than 0.1 μg L^{-1} in the final effluent. As mentioned above, only NP is subject to legislative limits and its effluent value is more than 20 times lower than its legislative limit (2 μg L^{-1}). These results show good agreement with a study of Clara *et al*. (2005), in which it is suggested that SRTs greater than 10 d reinforce the elimination of some biodegradable compounds like Ibuprofen and Bisphenol A. Moreover, Stasinakis *et al*. (2010), showed that a greater biodegradation of 4-n-Nonylphenol and Triclosan was achieved when the SRT was set to 20 days, comparing to 3 and 10 days. Last but not least, in relation to the RO implementation, a recent study appears to follow the same removal pattern of NP as the present one, since the compound never exceeded 120 ng L^{-1} , starting from an initial concentration of around 10^3 ng L^{-1} (Al-Rifai, et al., 2011). The removal of heavy metals was complete with only Pb and Mn being detected in a concentration level of less than 1 ppb and consequently the effluent stream fully met the legislation standards. Concerning the PPs, all of them were not detected in the MBR effluent with the exception of $CHCl_3$, the concentration of which rose from the influent to the MBR effluent and experienced only a slight decrease from the application of the RO. This deviant behavior is attributed to the volatile nature of $CHCl_3$, which might have caused a wrong influent concentration value. Still, all PPs had effluent values below of those set by the legislation. The concentrations of the influent and the MBR and RO effluent are presented in Table 4, while Table 5 compares the removal rates of heavy metals and PPs of this unit to other ones. From the MBR data provided in Table 5, it is evident that Cu and Mn removal rates are consistent with the ones in the cited studies. However, the removal rate of Zn agrees only with the one reported in the study of Mansell *et al*. (2004) and Gurung *et al*. (2016), but is less than half in comparison to other studies, while the removal of Fe is less than the ones observed in the rest of the studies. Another point worth mentioning is the low decrease rate of the $CHCl_3$ by the RO. A previous study found that RO can remove at least 80% of the inflowing $CHCl_3$ and also concluded that by increasing its concentration from 100 to 500 μg L^{-1} that rate decreased (Mazloomi, et al., 2009).

4. Conclusions

The results of this work, following the ones drawn from a previous study (Plevri, et al., 2017), lead to the conclusion

Table 2. MBR operational parameters

Parameter	Units	Value
Flow (Q)	$m^3 d^{-1}$	12
Hydraulic Retention Time (HRT)	h	3
Solid Retention Time (SRT)	d	20
Organic Loading (F M^{-1})	gCOD (gMLVSS d) $^{-1}$	0.38
Suspended Solids (MLSS)	g L^{-1}	9.2
Volatile Solids (MLVSS)	g L^{-1}	7.4
Sludge Removal (W)	L d^{-1}	84
Filtration Flux (J)	L m^{-2} h^{-1}	15-20
Filtration Flow (Q_{filtr})	L h^{-1}	500
Backflushing Flow (Q_{back})	L h^{-1}	1000

Table 3. Performance of the MBR-RO system for the treatment of municipal waste. The values presented refer to average values (range).

Parameter	MBR influent	MBR permeate	RO effluent
TSS (mg L^{-1})	106 (40-295)	≤5	≤5
VSS (mg L^{-1})	95 (34-240)	≤5	≤5
TDS (mg L^{-1})	Not Measured	672 (737-661)	179 (234-105)
COD (mg L^{-1})	342 (166-649)	25 (≤10-55)	≤10 (≤10-14)
CODs (mg L^{-1})	172 (80-241)	25 (≤10-55)	≤10 (≤10-14)
BOD_5 (mg L^{-1})	Not Measured	1.05 (0.2-2.42)	1.01 (0.2-2)
NH_4-N (mg L^{-1})	60 (20-79)	0.3 (0.09-0.7)	Not Measured
TN (mg L^{-1})	Not Measured	Not Measured	9 (6-17)
TP (mg L^{-1})	9 (6-11)	7 (6-8)	≤5
Conductivity (μS cm^{-1})	1500 (1250-1600)	1300 (1000-1530)	250 (160-650)
CL^- (mg L^{-1})	165 (133-213)	161 (129-199)	58 (16-136)
Turbidity (NTU)	Not Measured	0.06 (0.03-0.3)	-
TC [cfu $(100ml)^{-1}$]	$>10^7$	91 (7-470)	ND[1]
FC [cfu $(100ml)^{-1}$]	$>10^7$	5 (ND[1]-19)	ND[1]
EC [cfu $(100ml)^{-1}$]	$>10^7$	4 (ND[1]-19)	ND[1]

[1] Not Detected

that the installed MBR-RO pilot unit can produce water of excellent quality that meets the standards that are specified in the Greek National legislation regarding wastewater reuse for unrestricted irrigation and urban reuse. Such a dual membrane scheme in the context of a SM application has proven to be a viable solution for water reuse in combination with fresh water saving in highly urbanized, space-limited environments. Adding to that, the fact that European regulations could evolve in the future with the addition of new compounds and the gradual decrease in EQS values highlights the importance of technologies, such as the MBR-RO one, that can meet those criteria.

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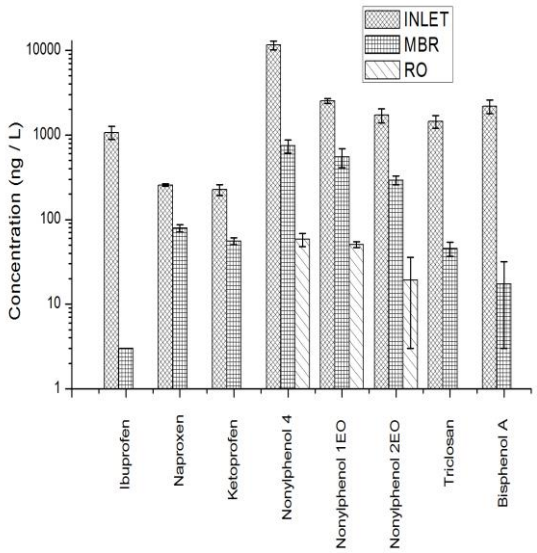

Figure 2. EDCs and NSAIDs average concentrations in the inlet, MBR permeate and RO effluent.

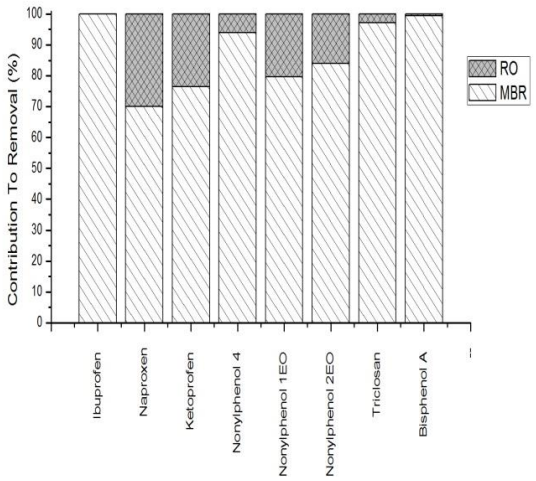

Figure 3. Contribution of MBR and RO to the total removal of EDC's and NSAIDs

Table 4. Concentrations of detected PPs and heavy metals in the influent MBR permeate and RO effluent streams (in μg L^{-1}).

Substance	Wastewater	MBR permeate	RO effluent	Legislation Limit
Al	480	120	-	5000
Cu	31	5	ND[1]	200
Fe	770	310	ND[1]	3000
Li	5	4	ND[1]	2500
Mn	42	6	0.39	200
Ni	5	<5	ND[1]	200
Pb	6	<5	<2.4	100
V	1	<1	ND[1]	100
Zn	110	64	ND[1]	2000
$CHCl_3$	0.18	0.27	0.23	2.5
TCE	0.23	ND[1]	ND[1]	10
PCE	0.14	ND[1]	ND[1]	10
C_6H_6	0.1	ND[1]	ND[1]	5

[1] Not Detected

Table 5. Comparison of MBR, RO and combined MBR-RO removal rates for selected metlas and PPs

MBR	RO	MBR-RO	Reference
Cu(84%), Al(75%), Fe(60%),Li(20%), Mn(86%), Zn(42%), TCE(>99.9%),PCE(>99.9%), C_6H_6(>99.9%)	Cu(>99.9%), Fe(>99.9%),Li(>99.9%), Mn(93.5%), Zn(>99.9%), C_6H_6(>99.9%) $CHCl_3$(15%)	Cu(>99.9%), Al(>75%),Fe(>99.9%),Li(>99.9%), Mn(99%), Zn(>99.9%), TCE(>99.9%),PCE(>99.9%), C_6H_6(>99.9%)	Present study
Cu(90%), Fe(85%), Mn(82%), Zn(75%)	-	Cu(>97.1%), Fe(>99.3%), Mn(>99.1%), Zn(>99.8%)	Malamis *et al*., 2011
Cu(95%), Zn(94%), Fe(97%)	-	-	Fatone *et al*., 2006

Cu(85%) [1], Zn(93%) [1], Fe(90%) [1], Al(94%) [1]	-	-	Carletti *et al*., 2008
Cu(31%) [1], Zn(60%) [1],	-	-	Gurung *et al*., 2016
Cu(>81%), Fe(>88%), Mn(>54%), Zn(26%), PCE(>99.9%), $CHCl_3$(>99.9)	-	-	Mansell *et al*.,2004 [2]

[1] Refer to average value, [2] Metal data retrieved indirectly through Conklin *et al* (2007), who used the raw data to extract the removal rates. Values refer to a Zenon pilot unit, [3] Values refer to case B, which treats only municipal wastewater with similar consistency with this study's influent

References

Akpor, O. B., Ohiobor, G. O., and Olaolu, T. D. (2014). Heavy Metal Pollutants in Wastewater Effluents: Sources, Effects and Remediation. *Advances in Bioscience and Bioengineering*, **2**, 37-43.

Al-Rifai, J., Khabbaz, H., and Schafer, A. (2011). Removal of pharmaceuticals and endocrine disrupting compounds in a water recycling process using reverse osmosis systems. *Seperation and Purification Technology*, **77**, 60-67.

American Puplic Health Association. (2012). *Standard Methods for Examination of Waters and Wastewaters* (22nd Ed ed.). Washington D.C.

Carletti, G., Fatone, F., Bolzonella, D., and Cecchi, F. (2008). Occurrence and fate of heavy metals in large wastewater treatment plants treating municipal and industrial wastewaters. *Water Science and Technology*, **57**,1329-1336.

Clara, M., Kreuzinger, N., Strenn, B., Gans, O., and Kroiss, H. (2005). The solids retention time-a suitable design parameter to evaluate the capacity of wastewater plants to remove micropollutants. *Water Research*, **39**, 97-106.

Comerton, A., Andrews, R., & Bagley, D. (2005). Evaluation of an MBR-RO system to produce high quality reuse water. *Water Research*, **39**, 3982-3990.

Fatone, F., Battistoni, P., Pavan, P., and Cecchi, F. (2006). Application of a membrane bioreactor for the treatment of low loaded domestic wastewater for water-reuse. *Water Science Technology*, **53**, 111-121.

Gao, D., Li, Z., Guan, J., and Liang, H. (2017). Seasonal variations in the concentration and removal of nonyphenol ethoxylates from the wastewater of a sewage treatment plant. *Journal of Environmental Sciences*, **54**, 217-223.

Gasperi, J., Garnaud, S., Rocher, V., and Moilleron, R. (2008). Priority pollutants in wastewater and combined sewer overflow. *Science of the Total Environment*, **407**, 263-272.

Gasperi, J., Garnaud, S., Rocher, V., and Moilleron, R. (2009). Priority pollutants in surface waters and settleable particles within a densely urbanised area:Case studies of Paris. *Science of the Total Environment*, **407**,.2900-2908.

Gulyas, G., Pitas, V., Fazekas, B., and Karpati, A. (2015). Heavy metal Balance in a communal wastewater treatment plant. *Hungarian Journal of Industry and Chemistry*, **43**, 1-5.

Gurung, K., Ncibi, M., and Sillanpaa, M. (2017). Assessing membrane fouling and the performance of pilot-scale membrane bioreactor (MBR) to treat real municipal wastewater during winter season in Nordic regions. *Science of the Total Environment*, **579**, 1289-1297.

Herojeet, R., Rishi, M., and Kishore, N. (2015). Integrated approach of heavy metal pollution indices and complexity quantification using chemometric models in the Sirsa Basin, Nalagarh valley, Himachal Pradesh, India. *Chinese Journal of Geochemistry*, **34**, 620-633.

Karvelas, M., Katsoyiannis, A., and Samara, C. (2003). Occurrence and fate of heavy metals in the wastewater treatment process. *Chemosphere* **53**, 1201-1210.

Kislenko, K., Dudutyte, Z., and Ruut, J. (2011). *Baltic Actions for Reduction of Pollution of the Baltic Sea from Priority Hazardous Substances.* Life+.

Lohami, M. B., Singh, A., Rupainwar, D. C., and Dhar, D. N. (2008). Seasonal varations of heavy metal contamination in river Gomti of Lucknow city region. *Environmental Monitoring and Assessment*, **147**, 253-263.

Malamis, S., Katsou, E., Takopoulos, K., Dmetriou, P., and Loizidou, M. (2012). Assessment of metal removal, biomass activity and RO concentrate treatment in an MBR-RO system. *Journal of Hazardous Materials*, **209-210**, 1-8.

Mansell, B., Kuo, J., Tang C, Thompson , S., Jackson, J., Garcia , A., and Stahl, J. (2004). Comparison of Two Membrane Bioreactors and an Activated Sludge Plant with Dual-Media Filtration : Nutrient and Priority Pollutant Removals. *Proceedings of the Water Environment Federation*, **20**, 749-761.

Mazloomi, S., Nabizadh, R., Nasseri, S., Naddafi, K., Nazmara, S., and Mahvi, A. (2009). Efficiensy of domestic reverse osmosis in removal of trihalomethanes from drinking water. *Iran. J. Environ. Health. Sci. Eng.*, **6**, 301-306.

Plevri, A., Mamais, D., Noutsopoulos, C., Makropoulos, C., Andreadakis, A., Rippis, K., Lytras, E and Lioumis, C. (2017). Promoting on-site urban wastewater reuse through MBR-RO treatment. *Desalinaton In proof corrected*.

Rule, K. L., Comper, S. D., Ross, D., Thornton, A., Makropoulos, C. K., and Rautiu, R. (2006). Diffuse sources of heavy metal entering an urbam wastewater catchment. *Chemosphere* **63**, 64-72.

Samaras, V., Thomaidis, N., Stasinakis, A., and Lekkas, T. (2011). An analytical method for the simultaneous trace determination of acidic pharmaceuticals and phenolic endocrine disrupting chemicals in wastewater and sewage sludge by gas chromatography- mass spectometry. *Analytical Bioanalytical Chem*, **399**, 2549-2561.

Sauve , S., and Desrosiers, M. (2014). A review of what is an emerging contaminant. *Chemistry Central*,**8** .

Soonthornnonda, P., & Christensen, E. R. (2008). Source apportionment of pollutants and flows of combined sewer wastewater. *Water Research*, **42**, 1989-1998.

Sorne, L., and Lagerkvist, R. (2002). Sources of heavy metals in urban wastewater in Stockholm. *The Science of the Total Environment*, **298**, 131-145.

Stasinakis, A. S., Kordoutis, C. I., Tsiouma, V. C., Gatidou, G., and Thomaidis, N. S. (2010). Removal of selected endocrine

disrupters in activated sludge systems: Effect of sludge retention time on their sorption and biodegradation. *Bioresource Technology*, **101**, p. 20902095.

Tiruneh, A. T., Fadiran, A. O., and Mitshali, J. S. (2014). Evaluation of the risk of heavy metals in sewage sludge intended for agricultural application in Swaziland. *International Journal of Environmental Sciences, 5*, pp. 197-216.

Ustin, G. E. (2009). Occurrence and removal of metals in urban wastewater treatment plants. *Journal of Hazardous Materials* **172**, 833-838.Wilkie, P. J., Hajimihalis, G., Koutoufides, P., and Connor, M. A. (1996). The contribution of domestic sources to levels of key organic and inorganic pollutants in sewage:the case of Melbourne, Australia. *Water Science and Technology*, **34**, 63-70.

Xiao, Y., Chen, T., Hu, T., Wang, D., Han, Y., Lin, Y., and Wang, X. (2014). Advanced treatment of semiconductor wastewater by combined MBR-RO technology. *Desalination*, **336**, 168-178.

Zhang, Q., Wang, F., Xue, C., Wang, C., and Chi, S. (2016). Comparative toxicity of nonylphenol,nonylphenol-4-ethoxylate and nonyphenol-10-ethoxylate to wheat seedlings. *Ecotoxicology and Environmental Safety* **131**, 7-13.