



Occurrence of UV filters, fragrances and organophosphate flame retardants in municipal WWTP effluents and their removal during membrane post-treatment



P. Krzeminski*, C. Schwermer, A. Wennberg, K. Langford, C. Vogelsang

Norwegian Institute for Water Research (NIVA), Gaustadalléen 21, N-0349, Oslo, Norway

HIGHLIGHTS

- Occurrence and removal of selected CECs from urban WWTPs effluents by UF, NF and RO.
- At least 75% removal of fragrances independent of the membrane used.
- The rejection rates of OPFRs strongly depended on membrane process applied.
- Membrane treatment reduced the potential risk in the receiving aquatic environment.

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ABSTRACT

Membrane filtration using ultrafiltration (UF), nanofiltration (NF) or reverse osmosis (RO) membranes was evaluated as an efficient effluent polishing step at municipal wastewater treatment plants (WWTPs) for the removal of selected contaminants of emerging concern and for improvement of water quality according to water reuse requirements. In samples collected at two largest WWTPs in Norway, 12 out of 14 selected personal care products and organophosphate flame retardants (OPFRs) were found above analytical detection limit. The highest concentrations were observed for BP3, OC (UV filters), HHCB, AHTN (fragrances), TCP and TBP (OPFRs), exceeding the predicted no-effect concentration for BP3 in one sample and AHTN in five samples. Independently of the membrane type used, membrane filtration effectively (>60%) removed BP3, UV-329, OC, HHCB, AHTN and DBPP. However, UF was insufficient (<20%) for removal of DEET, TCP and TCEP. UF was sufficient to remove 30–50% of COD, 80–95% of TP, up to 30% of TN and NH₄, and a min of 2 log reduction of *E. coli*. Water quality improved further with application of NF and RO. The results indicate that membrane filtration can be effective post-treatment to improve overall water quality and a measure to reduce potential risk in the receiving aquatic environment.

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1. Introduction

The global population growth and rapid urbanization results in increasing water demands and consumption, which subsequently results in a higher production of used and polluted water, i.e., wastewater, which may be harmful to the environment. For example, the expected population increase and concomitant increase in discharges from the main wastewater treatment plants (WWTP) VEAS and Bekkelaget in the Oslo area will add an extra burden to the already heavily loaded inner Oslofjord. In addition, water quality in the inner Oslofjord are of poor condition due to low oxygen levels

[1,2], especially in the Bunnefjorden where the WWTP Bekkelaget discharges influence the oxygen consumption.

Earlier estimates [2] have indicated that the discharges of nutrients (P and N) and organic matter from WWTPs is the single most important source of these contaminants and account for an estimated 50% of the oxygen consumption occurring below 20 m depth in the inner Oslofjord. Furthermore, contaminants of emerging concern (CECs) present in the treated effluent in the form of chemical and biological micropollutants may pose a risk to receiving waters and aquatic organisms. For example, some of the organophosphate flame retardants (OPFR) such as tricresylphosphate (TCP) and Tris(2-butoxyethyl)phosphate (TBEP), have been shown to be present in effluents from WWTPs at concentrations possibly posing a direct risk to the aquatic environment at the point of discharge in the Inner Oslofjord [3]. Several studies [4–11] have shown the

* Corresponding author.

E-mail address: pawel.krzeminski@niva.no (P. Krzeminski).

significance of WWTPs as important point sources for “old and new” organic and inorganic contaminants to the environment. In these studies, DEET, BP3, OC, HHCb, AHTN, TCPP, TCEP, DBPP, TBP (see [Tables 2](#) for compound description) were frequently found at elevated concentrations in effluents [8–11]. Moreover, the fate of these contaminants is highly dependent on the type of treatment applied at the specific WWTP [5–7]. Therefore, compounds which were expected to be present in elevated concentrations (e.g., DEET, BP3, OC, HHCb, AHTN, TCPP, TCEP, DBPP, TBP), potentially toxic to aquatic organisms (e.g., BP3, OC, TCEP), on national priority list of compounds to be eliminated from industrial use (i.e., TCEP) or ambiguous in terms of their removal (e.g., DEET, OC), were selected to investigate their occurrence and removal during membrane post-treatment step. In addition, consideration was given to the selection of the CECs from different functional groups and with different physico-chemical properties.

At present, CECs are not routinely monitored in WWTPs in Norway. Furthermore, no measures for the removal of those pollutants from the wastewater are currently in place, as there is no official obligation to this. In addition, conventional sewage treatment technologies are generally considered inadequate for the removal of many CECs as they are not designed to handle these pollutants. Considering all of the above, improving quality of the effluent produced at urban WWTPs is becoming a highly relevant issue. The removal of these compounds from the effluent is important because of their potential negative impact on the aquatic environment, human health and impairing quality of the water which could be potentially reused. Otherwise, the water quality in the effluent receiving water bodies will further deteriorate unless preventing measures are undertaken. In addition, in water reuse situation it is important to introduce a barrier that is capable of reducing the potential accumulation of contaminants. Hence, to prevent and reduce contamination of aquatic recipients by nutrients, organic matter and CECs, additional treatment solutions need to be considered. Reduction of the discharged pollution will help to at least maintain the current water conditions in the inner Oslofjord. One of the technologies suitable for the post-treatment of municipal effluent is membrane filtration.

In membrane filtration processes the removal of CECs typically occurs through the interactions between solutes and membranes: size exclusion, electrostatic repulsion, hydrophobic adsorption (on to the membrane) and partitioning [12,13]. These removal mechanisms are strongly influenced by a number of factors, like membrane process type, membrane characteristics, operating conditions and CECs characteristics and membrane fouling [14–16]. Among membrane filtration processes, low-pressure membranes processes such as microfiltration (MF) and ultrafiltration (UF) are generally less efficient for the removal of CECs as the membrane pore sizes are larger than the molecular sizes of CECs. Yet, CECs may be also removed through the adsorption on the membrane and interaction with organic matter in water. High-pressure membrane processes, such as nanofiltration (NF) and reverse osmosis (RO), are proven to be more suitable but at the expenses of higher costs mainly associated with energy demand and membrane replacement. On the other hand, the costs may be reduced through different options such as design solutions, hybrid systems, energy recovery systems, automation and control [17]. Until now, a number of studies have looked at the removal of various CECs [12–14,18–21]. However, despite increasing interest in membrane processes, membrane filtration of municipal effluent for removal of selected CECs has received little research attention [17,22,23].

The aim of this research is to evaluate the applicability of membrane filtration processes for the post-treatment of municipal WWTP effluent to improve water quality and removing emerging

chemical and biological (i.e., pathogenic bacteria) contaminants. The specific objectives of this study are:

- (1) to monitor the occurrence of selected organic micropollutants in the effluents of urban WWTPs,
- (2) to study removal efficacy of selected CECs in a bench-scale membrane filtration system,
- (3) to evaluate the efficiency of different membrane filtration processes (UF, NF and RO) for the post-treatment of effluent from municipal WWTPs, and
- (4) to highlight and evaluate reuse potential of the purified water.

2. Material and methods

Two sampling campaigns at two full-scale municipal WWTPs in Oslo area were carried out during 2014–2015. Each sampling campaign was followed by dedicated experimental campaign during which post-treatment of the collected effluent samples was carried out to elucidate the impact of membrane filtration on water quality improvements.

2.1. Sample collection

Samplings at VEAS and Bekkelaget WWTPs were carried out in October 2014 and February 2015, respectively. Detailed description of the full-scale municipal WWTPs consisting of mechanical, chemical and biological treatment steps can be found elsewhere [9,24].

At each facility, composite effluent samples, collected and stored cold over a 24 h period by means of the automatic sampling equipment used by the WWTPs for routine monitoring, were collected. Two 30–35L samples were collected at VEAS and four 10–15L samples were collected at Bekkelaget WWTPs during one week sampling. After collection, samples were immediately transported to the laboratory where experiments and analyses started as soon as possible to keep the characteristics of the samples relatively unchanged. The effluent samples were used as the influent in the membrane filtration experiments. From each membrane test samples of influent, permeate and concentrate were collected for further general water quality, chemical and bacterial analyses. Samples for chemical analyses were stored in organic free glass containers at -20°C . All samples for bacterial analyses were collected in sterile plastic bottles.

2.2. Membrane filtration tests

A custom-made bench scale membrane testing apparatus ([Fig. 1](#)) was used for evaluation of individual membranes. The tests were carried out on real effluent coming from VEAS and Bekkelaget WWTPs.

During each campaign, five different commercially available membranes in the UF, NF and RO range were tested on consecutive days. Small pieces of different flat sheet and spiral wound commercially available membranes were cut off for the testing in a filtration test cell. The effective membrane area used was of 99.4 cm^2 . The specifications of the individual membranes are listed in [Table 1](#).

Each membrane experiment was carried out with a new membrane and according to standardized protocol. The protocol consists of: membrane wetting in ultrapure water for minimum of 2 h to remove membrane coating; membrane wash according to manufacturers' recommendations which varies between water cleaning (GE, Toray) and alkaline wash (Alfa Laval, DOW); overnight membrane compaction with ultrapure water until a stable flux of permeate was obtained at the maximum pressure applied during the actual test; and actual filtrations for performance evaluation. Prior to the tests with WWTPs effluent, system was flushed for

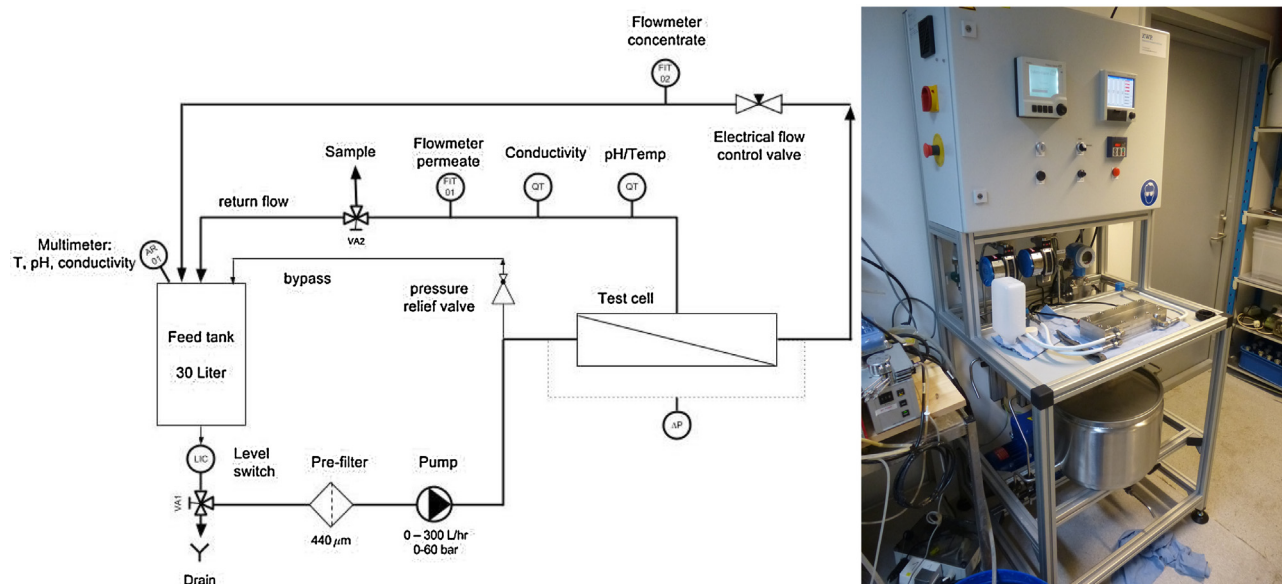


Fig. 1. Membrane testing apparatus.

Table 1
Description of evaluated membranes.

Membrane	Filtration spectrum	MWCO [Da]	Producer and brand name	Material
UF#1	UF	10 000	Alfa Laval, UFX-10pHt	Polysulphone permanently hydrophilic
UF#2	UF	1 000	GE, GE	Thin-film membrane
NF#1	NF	200–400	DOW, NF270	Polyamide thin-film composite
NF#2	NF	150	Toray, TM600	Piperazine polyamide composite
RO	RO	–	Alfa Laval, RO90	Thin-film composite on polyester support

MWCO – molecular weight cut off [Da]

30 min. All of the experiments were performed at ambient temperature of 21–22 °C and at constant pressure representative for each membrane type, i.e., 1–2 bar, 6–7 bar and 20–30 bar for UF, NF and RO membrane, respectively. Permeate collected within the first 1 h was discarded. The experiments were carried out until a total volume of 1.4 L of permeate was obtained.

2.3. Analyses

During each membrane evaluation test samples of influent, permeate and concentrate were collected for further analyses. Each sample was analysed for routine parameters (COD, TN, TP, NH₄) and selected contaminants of emerging concern: UV filters (BP-3, OC, EHCM, UV-329), fragrances (AHTN, HHCB), insect repellent (DEET) and organophosphate flame retardants (TBP, TBEP, TBPH, TCEP, TCPP).

The rejection (removal efficiency) values are defined as:

$$R_i = 1 - \frac{c_{i,p}}{c_{i,f}} \quad (1)$$

where R_i is the rejection of solute i (in %) and $c_{i,p}$ and $c_{i,f}$ are the concentrations of solute i in the permeate and the feed, respectively.

In order to avoid cross contamination, personnel did not use personal care products in the 24 h prior to sample collection and during testing period. Also disposable gloves were not used. Glass bottles used to collect samples for chemical analyses of CECs have gone through the heat treatment at 550 °C prior to use. Sterile plastic containers were used to collect samples for bacterial and antibiotic resistant gene analysis.

2.3.1. General water quality

Chemical oxygen demand (COD), ammonia (NH₄-N), total nitrogen (TN) and total phosphorus (TP) concentrations in the influent, permeate and concentrate were determined by photometrical methods with standard test kits (Hach-Lange, D). To carry out the reaction and to determine the concentration, a thermostat LT 100 (Dr. Lange) and a photometer DR 2800 (Hach-Lange) were used, respectively.

2.3.2. Chemical analyses

Concentrations of the selected UV filters (BP-3, OC, EHCM, UV-329), fragrances (AHTN, HHCB) and organophosphate flame retardants (TBP, TBEP, TBPH, TCEP, TCPP) were analysed in the membrane influent, permeate and concentrate.

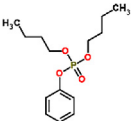
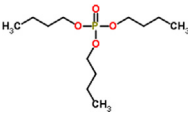
Samples were extracted onto solid phase extraction cartridges (Oasis HLB, 200 mg (Waters, Sweden)) after the addition of labelled internal standards. Cartridges were pre-washed with dichloromethane (DCM). Washing was followed by conditioning with methanol and water before sample extraction. Before elution, the cartridges were rinsed with ultrapure water and dried under vacuum. The analytes were eluted with DCM/ethyl acetate (50/50) and evaporated under nitrogen prior to analysis.

Analysis was performed on an Agilent gas chromatograph (GC) fitted with a 30 m × 0.25 mm, 0.25 μm film thickness DB-5MS column (Agilent Technologies) with helium carrier gas at a constant flow of 1 ml/min and a splitless injection at 250 °C. The initial oven temperature of 60 °C was held for 2 min, followed by an increase of 5 °C/min to 310 °C and held for 5 min. The GC was coupled to high-resolution time-of-flight mass spectrometer (GCT Premier, Waters Corp, Milford MA, USA) which was operated in full scan positive electron impact mode with a scan range of 50–800 m/z . The source

Table 2
Properties of UV filters, fragrances and organophosphate flame retardants; adopted from [9,11,25,26].

Group	Compound	Acronym	Structure ^a	CAS number	Log K _{OW}	Molecular weight [g/mol]	Function
Personal care products (PCPs)	N,N-diethyl-m-toluamide	DEET		134–62-3	2.4	191.27	Insect repellent
	benzophenone-3	BP3		131–57-7	3.8	228.24	Filter/ stabilizer
	octocrylene	OC		6197–30-4	7.3	361.48	Filter/ stabilizer
	ethylhexylmethoxycinnamate	EHMC		5466–77-3	5.8	289.39	Filter/ stabilizer
	2-(2H-Benzotriazol-2-yl)-4-(2,4,4-trimethyl-2-pentanyl)phenol	UV-329		3147–75-9	6.2	323.43	Stabilizer
	1,3,4,6,7,8-hexahydro-4,6,6,7,8,8-hexamethylcyclopenta-γ-2-benzopyran or Galaxolide®	HHCB		1222–05-5	6.3	258.40	Fragrance
	5-acetyl-3-isopropyl-1,1,2,6-tetramethylindane or Traseolide®	AITI		68140–48-7	5.36	258.40	Fragrance
	5-acetyl-1,1,2,3,3,6-hexamethylindan or Phantolide®	AHMI		15323–35-0	4.9	244.37	Fragrance
7-acetyl-1,1,3,4,4,6-hexamethyl-1,2,3,4-tetrahydronaphthalene or 1-(5,6,7,8-tetrahydro-3,5,5,6,8,8-hexamethyl-2-naphthyl)ethan-1-one or 6-acetyl-1,1,2,4,4,7-hexamethyltetraline or Tonalide®	AHTN		1506–02-1	5.4/5.7	258.40	Fragrance	
4-acetyl-6-tert-butyl-1,1-dimethylindan or Celestolide®	ADB1		13171–00-1	5.4	244.37	Fragrance	
Flame retardants	tris(2-chloro-isopropyl)phosphate	TCPP		13674–84-5	2.59/2.9	327.57	Flame retardant
	tris(2-chloroethyl)phosphate	TCEP		115–96-8	1.44/1.6	285.49	Flame retardant

Table 2 (Continued)

Group	Compound	Acronym	Structure ^a	CAS number	Log K _{ow}	Molecular weight [g/mol]	Function
	dibutylidiphenylphosphate	DBPP/DBPhP		2528–36-1	4.27/3.23	286.30	Flame retardant
	tributylphosphat	TBP		126–73-8	4.00	266.31	Flame retardant

^a <http://www.chemspider.com>.

temperature was 200 °C and the electron energy, 70 eV, and the resolution was 8500. The details related to the chemical analyses have been previously described in [3] and [9].

Properties of the investigated chemical contaminants are listed in Table 2.

2.3.3. Bacterial analyses

Number of *E. coli* and total coliform bacteria (TKB) were quantified by the most probable number method (MPN) with colilert-18 Quanti-Tray (IDEXX) according to ISO 9308-2:2012 [27].

3. Results and discussion

3.1. Occurrence of CECs

The presence of selected personal care products, namely, UV filters and stabilizers (BP3, OC, EHMC, UV-329), fragrances (HHCB, AITI, AHMI, AHTN, ADBI) and insect repellent (DEET), and organophosphorus flame retardants (TCPP, TCEP, DBPP and TBP) in the effluent of the municipal wastewater treatment plants (WWTPs) have been assessed. Details of the detected selected organic contaminants in the effluents from both WWTPs are presented in Fig. 2.

Total concentrations of UV filters in WWTPs effluent was between 300 and 1900 ng/L with BP3 and OC dominating in both of the WWTPs (Fig. 2). Together, BP3 and OC constituted more than 80% of the total UV filters concentration. Generally lower concentrations of the UV filters were observed during the previous studies carried out on 5 consecutive days at VEAS during 2013 year [9,11]. In 2013 concentrations of the BP3 and OC were in the range of 81–598 ng/L (median of 293 ng/L) and 181–538 ng/L (258 ng/L) compared to, respectively, 607–1099 ng/L (853 ng/L) and 310–763 (537 ng/L) in 2014. For EHMC and UV-329 levels below detection of 5 ng/L were observed in 2013, whereas in 2014 concentrations of <15–64 ng/L and 6–11 ng/L were measured for EHMC and UV-329.

Total concentrations of fragrances were between 1400 and 2100 ng/L with HHCB and AHTN concentrations predominant in the effluent streams with total presence above 98% of the analysed fragrances. The concentrations of HHCB and AHTN are higher compared to the results of the previous study on occurrence of fragrances in Norwegian WWTPs reported in 2011 [10] where concentrations in range of 412–673 ng/L for HHCB, and 50–64 ng/L for AHTN were reported for VEAS effluent. For Bekkelaget effluent, concentrations in range of 719–1259 ng/L for HHCB, and 67–87 ng/L for AHTN were measured during Norwegian screening programme in 2010 [10]. However, high concentrations in the range of 500–6400 ng/L for HHCB and 150–2700 ng/L for AHTN in the effluents of WWTPs in Austria, Germany, Spain, Switzerland, The Netherlands and UK were reported in 1997–2002 [25,28–33].

Total concentrations of OPFRs were between 2800 and 8250 ng/L with TCPP and TBP dominating the effluent streams and constituting to above 93% of the total OPFR. The concentrations are higher compared to the results reported in 2008 and 2011, especially TBP concentrations in Bekkelaget WWTP effluent which are 5 times higher compared to historical data. Previous studies [10] have reported concentrations in VEAS effluent of 800–960 ng/L and 72–88 ng/L for TCPP and TBP, respectively. The same study reported concentrations in Bekkelaget WWTP effluent of 720–1120 ng/L and 60–316 ng/L for TCPP and TBP, respectively [10]. In another study, concentrations of TCPP and TBP in the Bekkelaget WWTP effluent were reported as 2100 ng/L and 360 ng/L, respectively [34]. Interestingly, TCEP concentration of 1600–2200 ng/L observed in 2007 was highest among the years, i.e., 160–270 ng/L in 2010 and 320–400 ng/L in 2015, also when compared with the VEAS data: 88–148 ng/L in 2010 and 150–170 ng/L in 2014.

3.2. Removal of CECs by membrane filtration

The removal of CECs was evaluated during membrane filtration of the final effluent collected from VEAS and Bekkelaget WWTPs. The removal of TCEP is of special interest because it has been added to the national priority list of substances to be eliminated from industrial use. Results of the membrane post-treatment of effluent are presented in Figs. 3 and 4 and Table 3.

Overall, above 60% removal of BP3, UV-329, OC, UV-329, HHCB, AHTN and DBPP were achieved independently of the membrane used during the experiments.

UF membranes provided good removal in range of 75–99% (with many compounds below LoD) for the fragrances and in range of 60–99% for the UV filters with an exception for EHMC. Since the nominal pore sizes of the applied UF membranes are in range of 1000–10,000 Da, the size exclusion was not a major mechanism in removal of CECs having molecular sizes in range of 200–400 Da. Therefore, relatively good rejection is likely related to high logK_{ow} indicating high sorption potential, which in turn could suggest that these compounds were attached to the suspended particles and/or membrane through the hydrophobic adsorption mechanisms. However, UV filter with comparable logK_{ow} namely, EHMC did not follow the same trend. Poor removal (below 20%) of DEET and of most of OPFRs was observed when UF membranes were applied. Again higher rejections were observed for compounds with higher sorption capacity/logK_{ow}, i.e., TBP with rejection below 45% and DBPP with rejection about 75%.

The removal improved significantly when membranes of much 'tighter' structure, namely NF or RO, were used during the post-treatment. For example, DEET and OPFRs removal increased to 54–99%. Similar poor removal of DEET and TCEP by UF, and good to very good with RO, have been reported [20,21]. The physicochemical properties of DEET, TCEP and also TCPP indicate that sorption

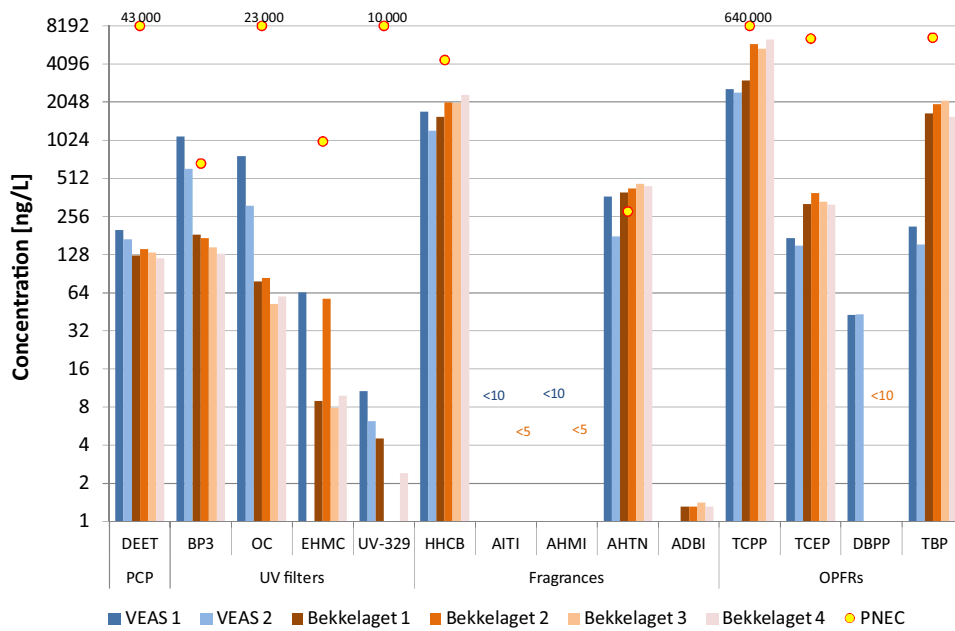


Fig. 2. Concentrations of UV filters, fragrances and organophosphate flame retardants detected in the effluent from wastewater treatment plants on two (VEAS) and four consecutive days (Bekkelaget). Note the logarithmic y-scale.

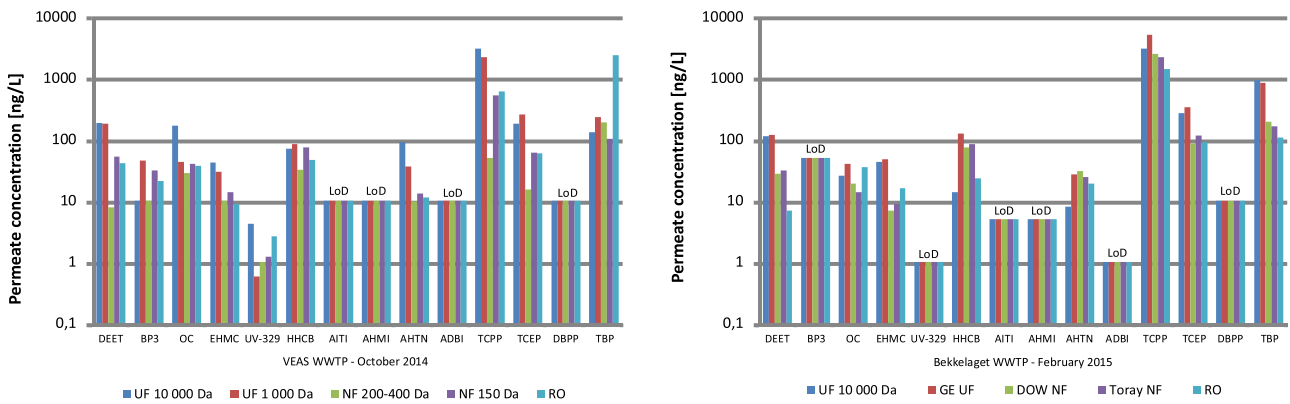


Fig. 3. Concentrations of UV filters, fragrances and organophosphate flame retardants (OPFR) in the membrane influent (WWTP effluent) and permeate. LoD indicates results below levels of detection.

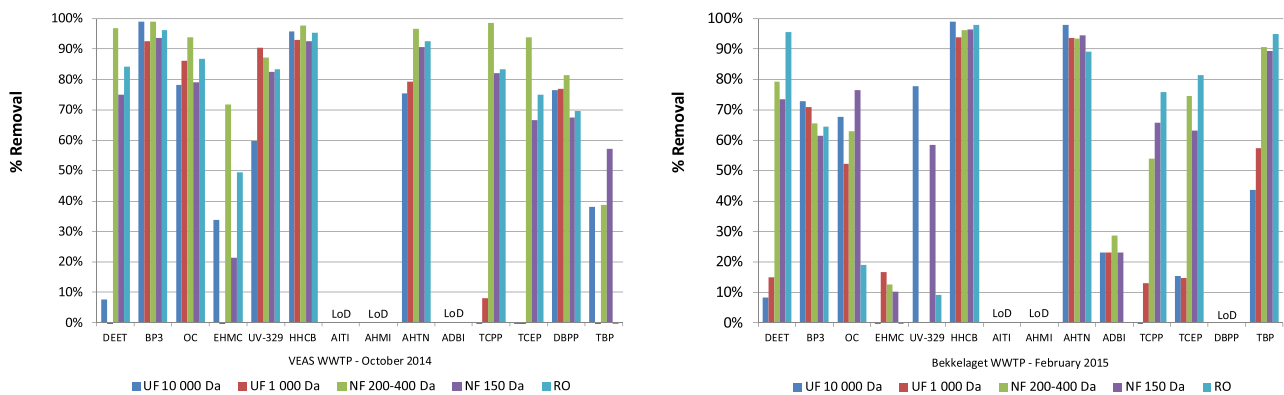


Fig. 4. Removal of UV filters, fragrances and organophosphate flame retardants (OPFR) achieved during polishing of the effluent. LoD indicates results below levels of detection.

is not a significant removal mechanism for these compounds. In addition, according to [35] hydrophilic compounds such as TCEP, TCP and DEET ($\log K_{ow} < 3$) do not adsorb to the membrane. Considering the MWCO of the NF (150–400 Da) and RO membranes

being in the range of 150–400 Da and ca. 100 Da, respectively, the improved removal may be attributed to size exclusion. In addition, electrostatic attraction or repulsion forces play a role in CECS rejection. Most of NF and RO membranes are negatively charged

Table 3
Concentrations of UV filters, fragrances and organophosphate flame retardants (OPFR) detected in the influent and permeate during membrane trials and associated removals of contaminants.

Compound	Group	UF#1, 10 000 Da			UF#2, 1000 Da			NF#1, 200-400 Da			NF#2 150 Da			RO		
		Influent [ng/L]	Permeate [ng/L]	Removal [%]	Influent [ng/L]	Permeate [ng/L]	Removal [%]	Influent [ng/L]	Permeate [ng/L]	Removal [%]	Influent [ng/L]	Permeate [ng/L]	Removal [%]	Influent [ng/L]	Permeate [ng/L]	Removal [%]
<i>VEAS 2014</i>																
DEET	Insect repellent	199	184	8 %	169	180	-6 %	257	8	97 %	209	52	75 %	260	41	84 %
BP3	UV filter	1099	<10	99 %	607	45	93 %	1051	<10	99 %	505	32	94 %	585	21	96 %
OC	UV filter	763	167	78 %	310	43	86 %	465	29	94 %	194	41	79 %	278	37	87 %
EHMC	UV filter	64	42	34 %	<15	30	N.M.	35	<10	72 %	18	14	21 %	17	9	49 %
UV-329	UV filter	11	4	60 %	6	1	90 %	8	<1	87 %	7	1	82 %	16	3	83 %
HHCB	Fragrance	1726	71	96 %	1220	84	93 %	1522	33	98 %	1012	75	93 %	1008	47	95 %
AITI	Fragrance	<10	<10	N.M.	<10	<10	N.M.	<10	<10	N.M.	<10	<10	N.M.	<10	<10	N.M.
AHMI	Fragrance	<10	<10	N.M.	<10	<10	N.M.	<10	<10	N.M.	<10	<10	N.M.	<10	<10	N.M.
AHTN	Fragrance	367	90	75 %	178	37	79 %	285	10	97 %	142	13	91 %	149	11	92 %
ADBI	Fragrance	<10	<10	N.M.	<10	<10	N.M.	<10	<10	N.M.	<10	<10	N.M.	<10	<10	N.M.
TCPP	OPFR	2575	3028	-18 %	2412	2218	8 %	3743	<50	99 %	2894	520	82 %	3665	612	83 %
TCEP	OPFR	172	182	-5 %	150	258	-72 %	248	15	94 %	185	62	67 %	241	60	75 %
DBPP	OPFR	43	<10	76 %	43	<10	77 %	54	<10	81 %	31	<10	67 %	33	<10	70 %
TBP	OPFR	214	133	38 %	154	232	-51 %	309	189	39 %	241	103	57 %	274	2376	-766 %
<i>Bekkelaget 2015</i>																
DEET	Insect repellent	125	115	8 %	142	121	15 %	134	28	79 %	120	32	74 %	156	<7	96 %
BP3	UV filter	185	<50	>73 %	172	<50	>71 %	145	<50	>65 %	130	<50	>62 %	141	<50	65 %
OC	UV filter	79	26	68 %	84	40	52 %	52	19	63 %	60	14	77 %	44	36	19 %
EHMC	UV filter	9	44	-391 %	58	48	17 %	8	7	13 %	10	9	10 %	12	16	-37 %
UV-329	UV filter	5	<1	>78 %	<1	<1	N.M.	<1	<1	N.M.	2	<1	>58 %	1	<1	N.M.
HHCB	Fragrance	1557	14	99 %	2046	124	94 %	2007	75	96 %	2350	85	96 %	1117	23	98 %
AITI	Fragrance	<5	<5	N.M.	<5	<5	N.M.	<5	<5	N.M.	<5	<5	N.M.	<5	<5	N.M.
AHMI	Fragrance	<5	<5	N.M.	<5	<5	N.M.	<5	<5	N.M.	<5	<5	N.M.	<5	<5	N.M.
AHTN	Fragrance	394	8	98 %	427	27	94 %	462	30	93 %	440	24	94 %	175	19	89 %
ADBI	Fragrance	1	<1	N.M.	1	<1	N.M.	1	<1	N.M.	1	<1	N.M.	<1	<1	N.M.
TCPP	OPFR	3027	3051	-1 %	5862	5103	13 %	5374	2470	54 %	6364	2182	66 %	5792	1402	76 %
TCEP	OPFR	322	273	15 %	393	335	15 %	335	86	74 %	318	117	63 %	490	91	81 %
DBPP	OPFR	<10	<10	N.M.	<10	<10	N.M.	<10	<10	N.M.	<10	<10	N.M.	<10	<10	N.M.
TBP	OPFR	1656	930	44 %	1965	836	57 %	2099	195	91 %	1553	164	89 %	2047	107	95 %

Legend: Orange colour indicates removal in range of 0-30%, blue in 30-70%, and green in 70-100%.

often providing improved rejection of negatively charged compounds due to electrostatic repulsion between negatively charged membrane and organic solute [20,36]. The fragrances removal above 89% observed for both NF and RO can be explained by the fact that molecular weight of fragrances is similar or higher than MWCO of the membranes and $\log K_{ow}$ is above 2 suggesting at least moderate to good removal due to steric exclusion and hydrophobic adsorption [14,37]. However, as pointed out by [37], the dominant mechanisms responsible for the CECs rejection depends on the properties of the target compound, membrane, operating conditions and membrane fouling. The key properties of the target compounds of influence on CECs rejection are molecular weight, molecular diameter, solutes dissociation constant (pK_a), hydrophilicity/hydrophobicity and diffusion coefficient. The rejection is also influenced by membrane properties such as pore size, MWCO, hydrophilicity/hydrophobicity, surface charge and morphology [15,37,38]. 19% removal of the UV filter OC during the RO experiments with the effluent of Bekkelaget WWTP is in contrast to 87% removal of OC from the VEAS effluent. In light of the OC molecular weight greater than membrane MWCO and $\log K_{ow} > 7$ high removal associated with steric exclusion and hydrophobic adsorption may be expected. Inconsistent results were also observed for the TBP and EHMC, both having $\log K_{ow} > 4$ and molecular weight in range of NF membranes. While expected rejection was observed for Bekkelaget when TBP removal varied from 44 to 57% for UF, around 90% for NF, up to 95% for RO membranes. Whereas for VEAS, TBP removal varied between 38% and 58% but also included two cases of increased concentration after membrane treatment possibly due to sample contamination. Also EHMC removal varied greatly between the experiments probably due to the increased uncertainty of quantification close to limits of EHMC detection (5–15 ng/L depending on the water matrix of the sample) and/or due to possible sample

contamination. The concentrations of AITI, AHMI and ADBI in the effluents were below the detection limit.

For most of the investigated CECs, NF offers similar removal efficiency as RO but at lower operating pressures resulting in lower energy demand [39]. Taking into account higher flux rates in NF and the fact that RO does not provide complete removal of CECs, NF seems to be more cost-effective barrier for CECs [12,19]. Nevertheless, additional treatment such as ozonation, adsorption onto activated carbon, advanced oxidation processes, may be needed to provide an absolute barrier for CECs. Ozonation and AOPs demonstrates high degradation rates and non-selectivity while simultaneously providing a disinfection step [13]. However, there is a need to evaluate the risk associated with the parent compounds, conjugates and by-products [40]. In addition, ozone efficiency is affected by DOC content so higher ozone dose may be required for elevated DOC levels. [41] observed 50–80% DEET removal, whereas [42] reported 50% removal of AHTN, 93% of HHCB, 27% of EHMC and <20% of OC at 5 mg/L O_3 dosage. Also activated carbon efficacy decreases under the presence of natural organic matter and CECs removal is dosage dependent. For example, [21,43] reported ca. $50 \pm 20\%$ removal of DEET, TCEP and HHCB with 5 mg/L dosage of powder activated carbon (PAC), whereas [42] reported 67% and 79% removal of AHTN and HHCB at a dose of 1.3 g/L. The same study, also obtained 91%, 92% and 95% removal of EHMC, OC and BP3 with low flow granular activated carbon (GAC) column. Therefore, PAC dose and GAC regeneration/replacement can be critical for activated carbon based processes [21].

3.3. Risk assessment of CECs

The concentrations of the contaminants measured during this campaign were compared with the published [9,44–50] predicted no effect concentrations (PNEC) to assess potential risk to the

Table 4

Predicted no effect concentrations (PNEC), predicted environmental concentrations (PEC) in the receiving water bodies based on the maximum and median values detected in effluent of Norwegian WWTPs, and calculated PEC/PNEC values for selected chemical contaminants. A general dilution factor of 10 was used for the water recipients to predict the PEC values.

Compound	WWTP _{effluent} concentrations [ng/L]	PNEC _{marine water} [ng/L]	PEC _{effluent,max} [ng/L]	PEC/PNEC [-]	PEC _{effluent,median} [ng/L]	PEC/PNEC [-]
DEET	120–199	43 000 ^a	20	0.0005	14	0.0003
UV filters						
BP3	130–1099	670	110	0.16	18	0.03
OC	52–763	23 000	76	0.003	8	0.0004
EHMC	8–64	1 000	6	0.006	1	0.001
UV-329	<1–11	10 000	1	0.0001	1	0.0001
Fragrances						
HHCB	1220–2350	4 400 ^a	235	0.05	187	0.04
AITI	<10	N.A.	LoD	N.A.	LoD	N.A.
AHMI	<10	N.A.	LoD	N.A.	LoD	N.A.
AHTN	178–462	280 ^a	46	0.17	41	0.15
ADBI	1.3–1.4	N.A.	LoD	N.A.	0.13	N.A.
Organophosphorus flame retardants (OPFRs)						
TCPP	2412–6364	640 000 ^a	636	0.001	420	0.001
TCEP	150–393	6 500 ^a	39	0.006	32	0.005
DBPP	<10–43	N.A.	4	N.A.	4	N.A.
TBP	154–2099	6 600	210	0.03	160	0.02

^a Fresh water; LoD – below level of detection; N.A. – not available.

environment. None of the considered contaminants exceeded the predicted environmental concentrations (PEC) to the predicted no effect concentrations ratio (PEC/PNEC) of 1 which would indicate potential risk for the local aquatic environment from the particular contaminant Table 4.

The effluent concentrations of selected UV filters observed in this study were well below the PNEC_{WWTP} for BP3, OC, EHMC and UV-329 indicating little risk to the WWTP microorganisms from the levels measured in the final effluent. The median concentrations of BP3, OC, EHMC and UV-329 are also below the PNEC for freshwater and marine water, except the concentrations of BP3 at VEAS WWTP (853 ng/L) which exceeds PNEC levels of 670 ng/L for marine water. The effluent concentrations of selected fragrances were also well below PNEC_{WWTP} for both HHCB and AHTN indicating little risk to the WWTP microorganisms from the levels of HHCB and AHTN measured in the final effluent. The median effluent concentrations of HHCB are also below the PNEC_{water} of 4400 ng/L. However, the median effluent concentrations of AHTN are at or above the PNEC_{water} of 280 ng/L indicating potential to pose a risk for the receiving environment. PNEC data are not available for AITI, AHMI and ADBI. Therefore, a dedicated screening campaign and more thorough risk assessment are needed to confirm these finding and to determine if the observed results are part of an isolated event or a general trend. The observed effluent concentrations of selected OPFRs were well below PNEC_{WWTP} for TCEP and TCPP indicating little risk to the WWTP microorganisms from the levels of TCEP and TCPP measured in the final effluent. The median effluent concentrations of TCEP, TBP and TCPP are also below the PNEC for fresh- and/or marine water of 6500 ng/L, 66,000/6600 ng/L and 640,000 ng/L, respectively. Therefore, since the levels determined in the effluent are below the PNEC levels, the undiluted effluent poses little risk for the receiving environment. PNEC data are not available for DBPP.

In conclusion, the levels of personal care products and OPFRs found in the effluents of the two largest Norwegian WWTPs are below the predicted no effect concentrations (PNEC) for selected compounds indicating little to no risk for the microbial communities within the WWTPs and, in most cases, also for the receiving water bodies. In case of BP3 and AHTN, the maximal detected concentrations were above the PNEC values for the receiving water bodies. This suggests that BP3 and AHTN may under certain low dilution conditions at the discharge points pose a risk to receiving

water body. However, after the additional treatment of the municipal effluents none of the analysed contaminants was present in the membrane polished effluent (i.e., permeate) in concentrations exceeding the PNEC levels for fresh- or marine water. Membrane filtration has been successful in effective reduction of BP3 and AHTN concentrations and producing water with BP3 and AHTN concentrations below the PNEC levels. All of five tested membranes applied during the experiments were able to sufficiently remove BP3 and AHTN. Therefore, membrane filtration can be considered as an effective post-treatment and a measure in reducing the potential risk for receiving environment.

3.4. Membrane post-treatment for water reuse purposes

3.4.1. Impact on general water quality parameters

The impact of post-treatment of the final effluent from VEAS and Bekkelaget WWTPs was assessed during experimental studies. To this end, effluent samples from each WWTP were subjected to membrane filtration with five different commercially available membranes in the UF-RO range. The results of the post-treatment performance are presented in Table 5 and Fig. 5. Bacterial results are based on the colilert most probable number (MPN) method.

The VEAS effluent sample was characterized by higher pollutants concentrations compared to Bekkelaget effluent, except for TP which was 50% lower. Despite the observed differences between the feed water, the UF membranes provided mostly comparable results in terms of achieved removals (Table 6). The removals were more variable for the NF#1, NF#2 and RO membranes, especially for the COD and NH₄, and ranged between 40 and 90% and 40–70%, respectively. This is likely due to different separation mechanism governing UF (primarily size-exclusion) and NF-RO (interactions between different transport processes namely diffusion, electromigration and convection) membranes. Removal of phosphorous varied between 83 and 99% and in general improved with membrane filtration range (i.e., UF < NF < RO). However, since all five membranes were able to remove TP to the concentration below the detection limit of 0.05 mg/L in the polished effluent, estimated TP removal performance is associated with some degree of uncertainty.

Contrary to UF#1 and NF#2, the NF#1 membrane did not provide sufficient reduction of pathogenic bacteria in case of Bekkelaget effluent. The removal of *E. coli* and TKB was not effective

Table 5
Concentrations and removal efficiency of COD, TP, TN, NH₄, *E.coli* and total coliform bacteria (TKB) in the WWTPs effluent and in the membrane post-treated effluents.

Membrane	Parameter	VEAS 2014			Bekkelaget 2015		
		WWTP effluent	Polished effluent	Removal [%]	WWTP effluent	Polished effluent	Removal [%]
UF#1 10 000 Da	COD [mg/L]	49.3	31.8	36	25.3	15.8	38
	TP [mg/L]	0.076	<0.05	87	0.162	<0.05	83
	TN [mg/L]	11.53	10.6	8	6.1	5.0	17
	NH ₄ [mg/L]	5.3	4.8	9	0.5	0.4	7
	<i>E.coli</i> [MPN/100 ml]	12590	1.0	99.99	9690	1.0	99.99
	TKB [MPN/100 ml]	81640	2.0	99.99	28820	12.1	99.96
UF#2 1000 Da	COD [mg/L]	45.3	23.6	48	23.5	10.8	54
	TP [mg/L]	0.080	<0.05	90	0.1	<0.05	95
	TN [mg/L]	10.77	8.5	21	6.9	5.2	25
	NH ₄ [mg/L]	4.8	3.4	30	0.1	0.0	29
	<i>E.coli</i> [MPN/100 ml]	N.M.	N.M.	N.M.	201100	59	99.97
	TKB [MPN/100 ml]	N.M.	N.M.	N.M.	345100	345	99.90
NF#1 200–400 Da	COD [mg/L]	39.5	20.1	49	20.4	6.0	70
	TP [mg/L]	0.079	<0.05	91	0.1	<0.05	95
	TN [mg/L]	11.20	7.9	30	6.1	5.1	17
	NH ₄ [mg/L]	5.0	2.4	52	0.0	0.0	66
	<i>E.coli</i> [MPN/100 ml]	N.M.	N.M.	N.M.	7380	228	96.9
	TKB [MPN/100 ml]	N.M.	N.M.	N.M.	26550	921	96.5
NF#2 150 Da	COD [mg/L]	36.8	21.1	43	27.4	3.4	88
	TP [mg/L]	0.062	<0.05	99	0.1	<0.05	99
	TN [mg/L]	11.30	9.6	15	5.9	4.4	26
	NH ₄ [mg/L]	5.0	4.0	21	0.1	0.1	16
	<i>E.coli</i> [MPN/100 ml]	N.M.	N.M.	N.M.	16130	1.0	99.99
	TKB [MPN/100 ml]	N.M.	N.M.	N.M.	41950	25.6	99.94
RO	COD [mg/L]	42.9	20.4	52	26.5	1.8	93
	TP [mg/L]	0.056	<0.05	80	0.1	<0.05	97
	TN [mg/L]	11.10	3.8	66	4.1	0.9	79
	NH ₄ [mg/L]	4.9	1.4	71	0.1	0.1	39

N.M. – not measured.

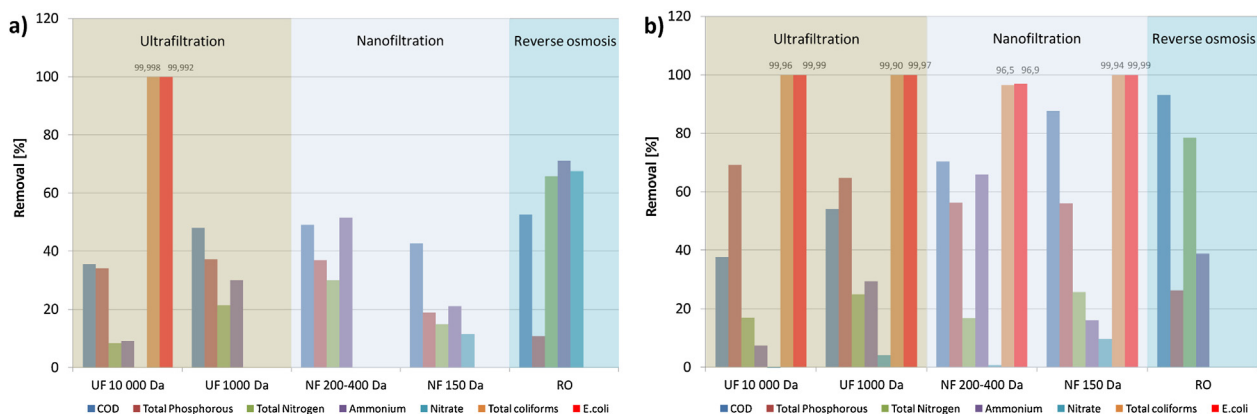


Fig. 5. Removal of COD, TP, TN, NH₄, *E. coli* and TKB during post-treatment of effluent from municipal WWTPs: a) VEAS 2014 and b) Bekkelaget 2015.

enough with removal of respectively, 96.9% and 96.5%, which corresponds to app. 1.5 log reductions. The removal of *E. coli* and TKB was not investigated for the RO membrane.

3.4.2. Water reuse potential

Depending on the membrane used water of different quality can be produced enabling fit-for-purpose approach. For example, based on NH₄ content, the UF post-treated water could be used for private and urban irrigation, while RO polished water could be used for environmental use, in aquaculture or even for industrial cooling (Table 7).

However, the reclaimed water would need to be analysed in a more comprehensive way beyond capabilities of this project by additional microbiological and chemical parameters in order to determine the permitted application of the reclaimed water. Example detailed list of the parameters which needs to be assessed can be found elsewhere [51].

At the same time, it is important to remember that different reuse applications will have different restrictions and need different treatment targets. Assuming the other parameters would not exceed their respective limits, the water reclaimed during the post-treatment could be applicable for private use in the households (e.g., house garden irrigation, toilet flushing, car washing), non-potable urban use (e.g., street cleaning, blue-green infrastructure) and irrigation of parks, gardens or golf courses. Furthermore, with the improved removal of nitrogen compounds the reclaimed water might also be permitted for environmental use or in the aquaculture. On the other hand, water used for irrigation would benefit from controlled levels of ammonia and *ortho*-phosphate, which may be achieved with membrane filtration.

The observed water quality improvement creates a good basis for potential water reuse in different not too demanding applications. This highlights the potential of using treated wastewater for

Table 6Concentrations and removal of COD, TP, TN, NH₄, *E.coli* and TKB achieved during post-treatment of WWTPs effluent classified by the membrane filtration range.

Membrane	Parameter	WWTP effluent	Polished effluent	Removal [%]	Removal [%]
UF	COD [mg/L]	24-49	11-32	36-54	30-50
	TP [mg/L]	0.08-0.16	<0.05	83-95	80-95
	TN [mg/L]	6-12	5.0-10.6	8-25	<25
	NH ₄ [mg/L]	0.1-5.3	0.0-4.8	7-30	<30
	<i>E.coli</i> [MPN/100ml]	9 690-201 100	1-59	99.97-99.99	>3 log
	TKB [MPN/100ml]	28 820-345 100	2-345	99.90-99.99	>3 log
NF	COD [mg/L]	20-40	3-21	43-70	40-70
	TP [mg/L]	0.06-0.12	<0.05	91-99	90-99
	TN [mg/L]	6-11	4.4-9.6	15-30	<30
	NH ₄ [mg/L]	0.1-5.0	0.0-4.0	21-66	20-60
	<i>E.coli</i> [MPN/100ml]	7 380-16 130	1-228	96.9-99.99	1.5-3 log
	TKB [MPN/100ml]	26 550-41 950	26-921	96.5-99.94	1.5-3 log
RO	COD [mg/L]	27-43	2-20	52-93	50-90
	TP [mg/L]	0.06-0.12	<0.05	80-97	80-97
	TN [mg/L]	4-11	0.9-3.8	66-79	70-80
	NH ₄ [mg/L]	0.1-4.9	0.06-1.4	39-71	40-70

Legend: Orange colour indicates removal in range of 0-30%, blue in 30-70%, and green in 70-100%.

Table 7

Selected water quality criteria for reclaimed water reuse as proposed by [51].

Parameter	Application example			
	Private, urban, irrigation	Environmental use	Aquaculture	Industrial cooling
pH	6.0–9.5	6.0–9.5	6.0–9.5	7.0–8.5
Conductivity [μ S/cm]	3 000	3 000	3 000	
COD [mg/L]	100	70–100	70–100	70
TSS [mg/L]	10–20	10–20	10–20	10–20
TN [mg/L]	15–25	10–20	10–20	10
Ammonium-N [mg/L]	2–20	1.5	1.5	1.5
TP [mg/L]	2–5	0.2–1	0.2–1	0.2
Faecal coliforms [cfu/100 ml]	ND-<1 000	ND-<1 000	ND-<10 000	ND-<10 000
Total bacteria [cfu/mL]	<1 000-<10 000	<1000-<10 000	<100 000	<10 000

ND – not detected.

beneficial purposes which until now has been little exploited in Norway.

4. Conclusions

The occurrence and removal of selected CECs from municipal WWTP effluent during membrane post-treatment was studied. Based on the results discussed in the paper, the following conclusions can be drawn.

- UF was sufficient to remove about 30–50% of COD, 80–95% of TP, up to 30% of TN and NH₄, and a min of 2 log reduction of *E.coli*, providing water with COD of 10–15 mg/L, TP of <0.05 mg/L, TN of 5 mg/L and NH₄ of 0.4 mg/L. The general water quality improved further with application of NF and RO membranes.
- At least 60% removal of BP3, UV-329, OC, UV-329, HHCb, AHTN and DBPP was achieved independently of the membrane used during the experiments. The removal of DEET, TCP and TCEP was below 20% for UF membranes, and in range of 54–99% for NF and RO membranes. The concentrations of AITI, AHMI and ADBI in the effluent were below the detection limit.
- Detected concentrations of BP3 and AHTN were above the PNEC values indicating that, under certain low dilution conditions at the discharge points, BP3 and AHTN may pose a risk to receiving water body. Contaminants were present in the membrane polished effluent in concentrations far below the PNEC levels for fresh- or marine water.
- Membrane filtration provides sufficient removal of chemical contaminants and a potent hygienic barrier for bacteria.

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