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Quantification and modelling of organic micropollutant removal by reverse osmosis (RO) drinking water treatment

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ABSTRACT

Reverse osmosis (RO) is the most promising membrane technology in organic micropollutants (MPs) removal of drinking water treatment. For 78 MPs, passage and removal were evaluated with an ESPA3 RO membrane and the robustness of RO against MPs was studied. The MPs were classified according to their charge and hydrophobicity. The results showed that the size of neutral compounds was negatively correlated with their passage. This correlation was weaker for neutral hydrophobic MPs than neutral hydrophilic MPs. The lowest passage (0.2%–4%) was displayed by anionic MPs because of electrostatic repulsion between the negatively charged solute and negatively charged membrane surface. Cationic MPs showed a higher passage (around 0.4%–40%) due to electrostatic sorption and Donnan exclusion. The relationship between physical-chemical properties of MPs and their passage was evaluated by the one-way analysis of variance (ANOVA). We performed a qualitative analysis of variables using Principal Component Analysis (PCA) in order to examine the physical-chemical properties of compounds that affect the membrane removal of MPs. After analysis with Multiple Linear Regression (MLR), we concluded that properties such as molecular width, equivalent molecular width, pKa and solubility can be considered as significant descriptors for prediction of the membrane removal. The influence of feed water temperature on MPs passage was also assessed. The results revealed that a rise of water temperature from 5 to 19 °C, increases the average passage of MPs by 6.5%.

1. Introduction

Micropollutants (MPs) in aquatic environment and natural drinking water sources have recently received attention as an emerging challenge in the scientific community all around the world [1–4]. The monitoring of organic MPs in the drinking water treatment process should be considered as a precautionary measure to address the concerns over the potential risks of MPs to human health [5,6]. Drinking water utilities tend to apply advanced technologies in order to remove unwanted chemicals. Although the natural pre-treatments (e.g., riverbank filtration, RBF) are efficient ways used by many drinking water utilities, not all the MPs are eliminated by the natural treatments [7] and therefore additional treatment is always necessary. Based on a several researches concerning the removal of MPs, conventional drinking water treatment processes show limitations in removing MPs, thus the development and

implementation of additional (advanced) treatment steps like activated carbon, ozonation, ultraviolet light and membrane treatment are strongly suggested [8,9]. Moreover, measuring the removal rate of MPs is confirmed as a high-priority task in drinking water contamination by the European Federation of National Associations of Water and Wastewater Services (Eureau) [10]. Eureau suggests conducting further studies on development of quantitative structure activity relationships (QSARs) [11]. In recent years, some research has been conducted investigating Multiple Linear Regression models [12], artificial neural network models (ANN) [13,14], principal component analysis [12,15] and QSAR models [16–19] in order to predict the removal efficiencies of MPs by different water treatment processes. For example, some QSAR models were developed to predict the MPs' removal efficiency of the river-bank filtration process [7]. However, all these models were evaluated to predict the removal of neutral or anionic MPs.

Table 1
Selected compounds and their physical-chemical properties.

Compound	MW (Da)	MV[a] (m³/mol)	Solubility[b] (mg/L)	Polarizability[b] (cm³)	Charge[c]	Log K_{ow}[b]	pKa (pKb)[c]	Log D[b]	Length (nm)[d]	Width (nm)[e]	Depth (nm)[e]	Eqwidth (nm)[e]	Category[f]
1,4-Dioxane	88.11	88.5	130,640	8.57E−24	Neutral	−0.27	(−3.9)	−0.09	0.480	0.280	0.267	0.273	2
1H-Benzotriazole	119.10	88.3	7570	1.38E−23	Neutral	1.44	8.6	1.28	0.620	0.490	0.000	0.007	2
2,4-D	221	148.0	336.2	1.94E−23	Negative	2.80	−4.9	−0.83	0.970	0.530	0.180	0.309	3
4-Methyl-1H-benzotriazole	133	105.0	4030	1.57E−23	Neutral	1.70	9.15	1.80	0.620	0.570	0.180	0.320	2
5-Methyl-1H-benzotriazole	133	105.0	2400	1.57E−23	Neutral	1.70	8.85	0.90	0.660	0.500	0.180	0.300	2
Acesulfame-K	201	108.0	74,905	1.29E−23	Negative	−2.67	3.02	−3.21	0.720	0.520	0.320	0.408	3
Aniline	93.10	91.7	18,000	1.21E−23	Neutral	0.90	4.6	1.14	0.560	0.420	0.019	0.089	2
Atrazine	215.70	170.0	27.5	2.32E−23	Neutral	2.61	15.8	2.20	1.030	0.680	0.300	0.452	1
Barbital	184.20	162.0	3230	1.75E−23	Neutral	0.65	8.48	0.60	0.700	0.560	0.460	0.508	2
Bentazone	240.30	179.0	268.6	2.37E−23	Negative	2.34	3.7	−0.19	0.960	0.506	0.330	0.409	3
Benzene	78.11	89.4	4230	1.04E−23	Neutral	1.99	NA	1.97	0.504	0.460	0.0002	0.010	2
Bisphenol A	228.29	199.5	86.5	2.70E−23	Neutral	3.32	9.6	4.04	1.070	0.624	0.393	0.495	1
Bisphenol S	250.27	175.0	3518	2.51E−23	Neutral	1.65	8.2	1.97	0.970	0.536	0.366	0.443	2
Carbamazepine	236.27	186.5	152	2.76E−23	Neutral	2.45	15.96	2.77	1.010	0.709	0.311	0.470	1
Carbendazim	191.19	135.0	2441	2.09E−23	Neutral	1.52	9.7	1.80	1.100	0.519	0.181	0.306	2
Chloridazon	222.00	155.0	3585	2.30E−23	Neutral	1.10	−1.8	1.10	0.950	0.560	0.170	0.309	2
Chlortoluron	213.00	175.0	329.1	2.32E−23	Neutral	2.40	13.5	2.50	0.930	0.540	0.430	0.482	1
DEP	222.24	198.0	108.63	2.34E−23	Neutral	2.42	(−6.7)	2.69	0.940	0.617	0.350	0.465	1
Diatrizoate	613.92	234.0	107	3.98E−23	Negative	1.37	2.17	−0.63	0.960	0.710	0.302	0.463	3
Diatrizoic acid	613.90	234.0	107	3.98E−23	Negative	−1.28	2.17	−0.63	0.960	0.780	0.610	0.690	3
Diclofenac	294	207.0	4.47	3.03E−23	Negative	4.26	4.15	1.10	0.930	0.681	0.496	0.581	3
Diglyme	134.18	147.0	60,900	1.39E−23	Neutral	−0.36	(−3.7)	−0.32	1.110	0.213	0.181	0.196	2
Dimethenamid	275.80	231.0	247	2.92E−23	Neutral	2.15	16.73	2.92	0.910	0.655	0.382	0.500	1
Dimethomorph	387.90	315.0	5240	4.16E−23	Neutral	2.68	−0.46	3.28	1.300	1.040	0.410	0.653	1
Diuron	233.00	170.0	150.6	2.33E−23	Neutral	2.70	13.2	2.50	0.910	0.540	0.410	0.471	1
DNOC	198.13	128.0	678.4	1.83E−23	Negative	2.27	4.31	−0.49	0.690	0.639	0.181	0.340	3
Erythromycin	733.50	607.0	459	1.88E−23	Positive	3.06	9.69	1.69	1.260	1.044	0.841	0.937	4
Estrone	270.40	232.1	3.94	3.08E−23	Neutral	3.13	10.4	3.38	1.150	0.563	0.386	0.466	1
ETBE	102.18	134.0	32,950	1.25E−23	Neutral	1.92	NA	1.54	0.650	0.429	0.402	0.415	2
Furosemide	330.70	206.0	149.3	3.00E−23	Negative	2.03	4.25	−0.78	1.280	0.630	0.330	0.456	3
Gabapentin	171.20	162.0	4340	1.85E−23	Neutral	−1.10	4.63	−1.27	0.740	0.590	0.400	0.486	2
Guanylurea	102.00	56.6	19,339,070	8.49E−24	Positive	−3.60	13.62	−3.85	0.650	0.370	0.087	0.179	4
HFPO-DA	330.10	188.7	15.17	1.37E−23	Negative	3.66	3.8	1.34	0.850	0.450	0.320	0.379	3
HMMM	390.40	320.0	149.3	4.03E−23	Neutral	1.61	NA	1.54	1.320	0.980	0.850	0.913	2
Hydrochlorothiazide	297.70	176.0	1292	2.49E−23	Neutral	−0.07	9.09	−0.70	0.940	0.560	0.330	0.430	2
Ibuprofen	206.30	200.0	68.4	2.41E−23	Negative	3.97	4.8	1.70	1.060	0.472	0.338	0.399	3
Iopamidol	777.09	341.0	790	5.44E−23	Neutral	−2.42	11	−2.31	1.300	0.977	0.504	0.702	2
Isoproturon	206.00	196.0	143.8	2.50E−23	Neutral	2.90	13.8	2.30	1.057	0.520	0.410	0.462	1
MCPA	200.60	153.0	180.9	1.94E−23	Negative	3.25	3.13	−1.00	0.950	0.558	0.181	0.318	3
MCPP	196.68	170.0	841.35	2.12E−23	Positive	2.11	8.87	0.67	1.200	0.667	0.394	0.513	4
Melamine	126.10	75.9	257,100	1.32E−23	Positive	−1.37	(−9.56)	−2.34	0.620	0.600	0.013	0.088	4
Metamitron	202.24	150.0	8334	2.23E−23	Neutral	0.83	1.9	0.87	0.980	0.474	0.197	0.306	2
Metformin	129.00	101.0	1380	1.32E−23	Positive	−1.40	12.3	−3.80	0.710	0.430	0.360	0.393	4
Methenamine	140.20	98.2	3,457,320	1.55E−23	Neutral	−4.15	(−5.88)	0.38	0.470	0.420	0.390	0.405	2
Metoprolol	267.37	259.0	402	3.06E−23	Positive	1.88	14.09	−0.47	1.530	0.587	0.399	0.484	4
Metribuzin	214.29	164.0	1304	2.27E−23	Neutral	1.70	−2.46	1.50	0.930	0.563	0.408	0.479	2
MTBE	88.15	117.0	66,470	1.07E−23	Neutral	1.18	(−4.1)	1.18	0.530	0.400	0.337	0.367	2
Naphthalene-1,3,6-trisulfonic acid	368.00	192.0	18,780	2.99E−23	Negative	−1.90	NA	−6.30	1.021	0.840	0.290	0.494	3
Naphthalene-1,5-disulfonic acid	288.00	169.0	9195	2.58E−23	Negative	−0.90	NA	−4.60	0.870	0.720	0.270	0.441	3
NDMA	74.08	75.0	2360	7.36E−24	Neutral	−0.57	(−3.52)	0.04	0.420	0.364	0.181	0.257	2
Nicosulfuron	410.00	284.0	14.44	3.82E−23	Negative	0.01	4.5	−2.31	1.380	0.870	0.410	0.597	3
NMOR	116.12	87.3	305,655	1.10E−23	Neutral	−0.44	(−3.14)	−0.18	0.550	0.457	0.311	0.377	2

Table 1 (*continued*)

Compound	MW (Da)	MV[a] (m³/mol)	Solubility[b] (mg/L)	Polarizability[b] (cm³)	Charge[c]	Log K$_{ow}$[b]	pK$_a$ (pK$_b$)[c]	Log D[b]	Length (nm)[d]	Width (nm)[e]	Depth (nm)[e]	Eqwidth (nm)[e]	Category[f]
NTA	191.00	119.0	738,900	1.52E−23	Negative	−3.80	1.97	−5.92	0.800	0.480	0.300	0.379	3
Pentoxifylline	278.00	211.0	453.7	2.94E−23	Neutral	0.30	19.64	0.54	1.250	0.640	0.310	0.445	2
PFBA	214.00	127.0	765.7	9.19E−24	Negative	2.40	1.6	−1.20	0.600	0.290	0.270	0.280	3
PFBS	300.00	162.0	344	1.27E−23	Negative	2.40	−3.3	0.20	0.710	0.400	0.230	0.303	3
PFHA	314.00	182.0	15,700	1.31E−23	Negative	4.40	−0.16	0.15	0.800	0.330	0.230	0.275	3
PFHS	400.12	217.0	6.2	1.66E−23	Negative	4.34	0.14	0.15	0.800	0.460	0.370	0.413	3
PFNA	464.00	265.0	9500	1.90E−23	Negative	7.30	−0.21	2.84	1.300	0.300	0.230	0.263	3
PFOA	414.07	237.0	9500	1.70E−23	Negative	4.81	−4.2	1.60	1.170	0.283	0.227	0.253	3
PFOS	500.13	272.0	680	2.04E−23	Negative	4.49	0.1	0.66	1.200	0.353	0.262	0.304	3
Phenobarbital	232.20	188.0	1110	2.35E−23	Neutral	1.47	8.14	1.19	0.910	0.530	0.460	0.494	2
Pirimicarb	238.29	208.0	2700	2.62E−23	Neutral	1.70	(−4.99)	1.80	0.950	0.734	0.410	0.549	2
Propranolol	259.30	237.0	61.7	3.13E−23	Positive	3.48	14.09	1.15	1.300	0.670	0.450	0.549	4
Pyrazole	68.10	61.0	19,400	7.44E−24	Neutral	0.26	14.76	0.28	0.430	0.330	0.00012	0.006	2
S-Metolachlor	284.00	258.0	50.86	3.17E−23	Neutral	3.13	NA	3.22	0.860	0.820	0.650	0.730	1
Sotalol	293.81	220.0	782	2.86E−23	Positive	0.24	10.07	−1.63	1.120	0.920	0.412	0.616	4
Sucralose	397.60	235.0	22,700	3.20E−23	Neutral	−1.00	11.9	−0.17	1.070	0.700	0.530	0.609	2
Sulfamethoxazole	253.30	173.0	3942	2.42E−23	Negative	0.89	6.2	0.10	1.280	0.477	0.236	0.336	3
TBA	74.12	92.1	110,080	8.75E−24	Neutral	0.35	18.09	0.72	0.430	0.351	0.308	0.329	2
TCEP	285.49	205.0	877.9	2.24E−23	Neutral	1.44	(−9.1)	1.42	0.940	0.747	0.367	0.524	2
Terbuthylazine	229.70	186.0	9	2.50E−23	Positive	3.21	2	2.99	0.950	0.720	0.390	0.530	4
Tetraglyme	222.30	225.0	254,000	2.26E−23	Neutral	−1.03	(−3.4)	−0.33	1.820	0.210	0.180	0.194	2
TFA	114.00	72.6	1,483,280	5.29E−24	Negative	0.50	0.95	−2.62	0.300	0.280	0.210	0.242	3
Tiamulin	493.70	477.0	0.6957	5.58E−23	Positive	4.75	14.43	3.33	1.490	0.900	0.590	0.729	4
TPPO	278.30	236.0	62.76	3.29E−23	Neutral	2.83	NA	3.67	0.950	0.820	0.450	0.607	1
Tramadol	263.40	255.7	1151	3.09E−23	Positive	2.51	13.8	0.52	1.170	0.710	0.500	0.596	4
Triglyme	178.23	186.0	460,910	1.83E−23	Neutral	−0.76	(−3.5)	−0.38	1.500	0.223	0.181	0.201	2

[a] MV = molar volume; calculated with compTox chemical dashboard.

[b] Calculated with PubChem database and ChemSpider database.

[c] Calculated with Chemaxon software; when pKa (strongest acidic) was not available pKb (strongest basic) was considered.

[d] Calculated by Chem3D software.

[e] Calculated with HyperChem software.

[f] Properties category 1: neutral-hydrophobic MPs; category 2: neutral-hydrophilic MPs; category 3: anionic MPs; category 4: cationic MPs.

In this article we focus on reverse osmosis (RO) drinking water treatment which has been demonstrated to be an appropriate technology for removing MPs [17,19]. RO is a water purification process that uses the solution-diffusion model for transporting the organic solutes through osmotic membranes [20,21]. Many studies have investigated the separation mechanisms of MPs which are explained by the physical-chemical properties of solutes and membrane characteristics leading to the qualitative description of the removal efficiencies of MPs and prediction of the rejection of target compounds. As recently reviewed by Kim et al. [20], the removal of neutral MPs by RO membranes is governed by the hindrance and size exclusion. However, higher removal efficiencies were associated with the hydrophobic-hydrophobic interactions between compounds and membrane [22,23]. Mechanistic models used to describe removal of charged solutes are often represented by adsorption due to partition and electrostatic repulsion or attraction [24–27]. In general, the removal of MPs is mostly influenced by compounds properties, feed water characteristic and operating conditions [28,29,12,15].

Modelling the removal of MPs by RO is of great importance for improving membrane technologies and to predict rejection quantitatively. Several assessments have demonstrated that the MW and hydrophobicity of compounds can be useful predictors for the RO removal of non-charged and non-polar compounds [23,26,30,31]. On the other hand, some studies showed that the MW of a polar solute with hydrophilic characteristic is a rather poor predictor of their removal [32]. Other studies [14,18] displayed that although MW was a fundamental predictor for MPs removal, it was not as accurate as the geometrical descriptors of compounds. However, there is still a lack of studies concerning the influence of other physical-chemical properties on the removal of MPs by RO.

In association with compound's physical-chemical properties, the removal of MPs is also dependent on the membrane characteristics [18]. Verliefde et al. [25] presented a simplified rejection model for charged organic compounds in order to improve the understanding of removal mechanisms of negatively charged MPs. Their results showed that the membrane charge plays a significant role in removal of charged compounds [25]. The molecular weight cut-off (MWCO) is one of the membrane properties which usually refers to the MW of the molecule that is between 60 and 90% retained by the membrane [33]. In some studies, MWCO has been used for prediction of MPs rejection [18], while Kimura et al. [34] argued that MWCO might be a good semi-quantitative predictor for modelling the removal of MPs. Porosity of the membrane is another useful parameter which has been used to estimate compound separation and usually expressed as pore size in the membrane's upper layer [32]. Thus, it is evident that the membrane properties such as the MWCO and porosity of the membrane play an important role in compound's separation and removal efficiencies by an RO membrane.

These previous studies demonstrated that solute properties (e.g., MW, geometry, charge, hydrophobicity), membrane properties (e.g., MWCO, pore size, charge) as well as the feedwater composition (e.g., temperature, pH) need to be considered when estimating the removal of MPs by RO membranes. Therefore, any improvement of the understanding regarding the effect of different RO membrane properties on solutes transfer might be essential for the further development of predictive tools and robustness test.

In this study the removal and passage of MPs in a pilot-scale RO drinking water treatment concerning both charged and non-charged compounds were quantified. In addition to the mentioned solute physical-chemical properties, the effect of solubility, polarizability, and acid dissociation constant (pKa) of compounds on their removal by the ESPA3 RO membranes was assessed and modelled by a multivariate statistical technique. Moreover, to improve the description of the solute size's influence, the descriptors: molecular length (length), molecular depth (depth), molecular width (width), and equivalent molecular width (eqwidth) were considered besides the MW. To the best of our knowledge, the removal of charged (anionic and cationic) and uncharged compounds by ESPA3 RO membrane has been reported in only

very few studies [35], while even fewer are the studies in which the rejection efficiency has been modelled. Moreover, with this paper we report the removal of some MPs which have never been measured in RO filtration (e.g., nicosulfuron, naphthalene-1,3,6-trisulfonic acid and naphthalene-1,5-disulfonic acid). In addition, the effect of feed water temperature on the removal efficiency of MPs was investigated.

2. Materials and methods

2.1. MPs selection

The robustness of drinking water treatments has been studied for several years in the Netherlands and assesses the extent to which specific purification processes (or their combinations) form a barrier against old and emerging priority substances. This research is carried out in pilot plants, which have the same process set-ups as full-scale plants applied by drinking water companies in the Netherlands. In the pilot plants, MPs are dosed, and the removal is determined. The robustness research consists of a five-year cycle each time. The first cycle was carried out in 2006, the second cycle in 2012 and the third cycle of robustness research was carried out in 2017. All the dosing tests were also conducted at PWN water company, which provides for the drinking water supply in the province of North Holland. PWN tests were based on a RO pilot plant. In total 78 MPs were selected during these three cycles between 2006 and 2017 (Table 1).

In 2006, 35 MPs have been selected based on their detection in natural freshwater, their toxicological value, their interest for the water utilities and their variation in polarity, molecular size or molecular weight. In 2012, the study was conducted on a selection of 21 compounds (presence in freshwater resources, toxicity, etc.) and aimed to determine the removal of these substances through various purification techniques such as RO. Lastly in 2017, 22 MPs were selected based on their presence in the Association of River Waterworks database [36], in research reports [37–39], in the Water Framework Directive guideline [40,41] while also some substances were proposed by the Dutch drinking water utilities.

2.2. Sample analyses

In this study compounds used in the pilot installation were divided into three groups, based on their solubility, consisting of soluble group (1), poorly soluble group (2) and stock solutions were made accordingly. Stock solution (3) consisted of HFPO-DA (GenX) and volatile compounds which were added on the day of dosage. Stock solution (1) and (2) were prepared in 20 L stainless steel tanks which was filled with ~10 L of demineralized water. The weighted compounds were added sequentially and after that demineralized water was added to obtain a final volume of ~15 L. Stock solutions were stirred for 6 days at 35 °C. Stock solution (3) was prepared in a 20 L stainless steel tank filled with ~15 L of demineralized water. After addition of HFPO-DA (GenX), the solution was kept at room temperature under constant stirring for at least 12 h. In addition, all stock solutions were filtered through a 0.20 μm filter and demineralized water was added to 20 L by weight in jerry cans. Stock solutions were stored at 3 ± 2 °C until the day of dosage. Detailed information on preparation of the stock solutions for spiking has already been described and published in Brunner et al. [42]. The methods applied to monitor the chemical concentrations have been described in Supplementary materials Section 1.1.

2.3. RO pilot feed water

The feed water of the RO pilot was abstracted from the IJsselmeer lake and pre-treated with drum sieves, coagulation and sedimentation, rapid sand filter and ultrafiltration (UF). This pre-treatment is essential since IJsselmeer water has a very high fouling potential for UF-RO systems. Thus, the RO pilot was fed with UF effluent in which the MPs

Table 2
ESPA3 RO membrane characteristic.

Performance		Properties			
Salt removal[a] (%)	Permeate flux[a] (m³/day)	Surface area[a] (m²)	MWCO[b] (Da)	Pore size[c] (nm)	Zeta-potential[d] (mV)
98.5	11.4	7.9	<200	0.2–1	<−20

[a] Manufacturer data.

[b] MWCO: molecular weight cut-off; Yangali-Quintanilla et al. [13]; Dach [63].

[c] Kosutic et al. [80].

[d] Tu et al. [67].

were spiked from the stock solutions to the final target concentration. A stock solution was diluted with RO permeate in a tank of 600–700 L containing a stirrer. The flow through the RO pilot was 9.7 m³/h. More information about the feed water properties and a schematic representation of the pilot installation are provided in Supplementary material Sections 1.2 and 1.3, respectively.

2.4. Set-up pilot-scale installation

The RO installation consisted of a two-stage installation with a 4×2 pressure vessel circuit with 7 elements per pressure vessel. The polyamide-based RO membrane used in this study was Hydranautics ESPA3, 4040. The properties of the membrane are summarized in Table 2. The recovery and permeate flux of the filtration were 82% and 30 L m⁻² h⁻¹, respectively. The total contact time of the RO installation was low, and a dosing experiment could take at least 48 h. In a short-term dosing experiment, the removal by adsorption may be obtained instead of removal by membrane filtration [43]. Thus, to minimise the impact of adsorption on the removal of hydrophobic MPs and to obtain the steady-state rejection values, the tests were carried out for 3-4 days in replicate. A detailed description of the RO pilot is provided in the Supplementary material Section 1.3.

Also, the removal efficiency of 26 MPs (Table S9) at two different feed water temperatures were measured in order to focus on the effect of temperature on the removal of selected compounds. The water quality was similar during these studies on most standard parameters while the temperature was 5 °C during the first experiment and 19 °C during the second experiment.

2.5. MPs classification

The MPs were classified based on their charge and hydrophobicity into four categories (Table 1). The pH-dependent octanol-water distribution coefficient (log D_{OW}) was used to describe hydrophobicity. The compounds with log D (pH$_7$) ≥ 2 are referred to as hydrophobic and those with log D (pH$_7$) < 2 are hydrophilic [44,45]. Therefore, the compounds were categorized as: (1) neutral and hydrophobic (HP); (2) neutral and hydrophilic (HL); (3) anionic; (4) cationic.

Category 1 included twelve neutral HP MPs; Category 2 included thirty-one neutral HL MPs; category 3 included twenty-four anionic MPs and category 4 included eleven cationic MPs. The list of compounds and their classification in category 1-4 in Table 1 also show the physical-chemical properties of the investigated compounds such as MW, molar volume (MV), water solubility, polarizability, charge of compounds, water-octanol partition coefficient (log K_{ow}), log D, pKa, length, width, depth and eqwidth. When the experimental value was missing, log K_{ow} values were obtained from software KOWWIN [46]. Although log K_{ow} is used to describe solute hydrophobicity in many studies [12], the hydrophobicity/hydrophilicity of compounds was quantified also in terms

Table 3
Average removal (%) and range (n); the values under the detection limit are shown in bold.

Num.	Compound	n	Min	Max	Average removal %	Passage %
1	1,4-Dioxane	2	89.7	91.1	90.4	9.6
2	1H-Benzotriazole	2	8.2	27.7	15.6	84.4
3	2,4-D	4	99.6	99.8	99.7	0.3
4	4-Methyl-1H-benzotriazole	6	14.0	64.8	38.7	61.3
5	5-Methyl-1H-benzotriazole	6	8.7	45.7	24.4	75.6
6	Acesulfame-K	6	98.1	99.9	99.6	0.4
7	Aniline	2	47.3	70.0	59.1	40.9
8	Atrazine	6	99.1	99.7	99.4	0.6
9	Barbital	2	91.7	94.3	**93.1**	**6.9**
10	Bentazone	2	99.3	99.4	99.3	0.7
11	Benzene	2	8.3	21.7	13.7	86.3
12	Bisphenol A	2	98.5	98.9	98.7	1.3
13	Bisphenol S	6	97.1	99.7	98.5	1.5
14	Carbamazepine	8	97.1	99.9	98.9	1.1
15	Carbendazim	4	75.0	81.9	80.1	19.9
16	Chloridazon	4	83.7	95.6	91.3	8.7
17	Chlortoluron	4	85.3	95.9	92.0	8.0
18	DEP	2	94.9	95.8	95.3	4.7
19	Diatrizoate	2	99.2	99.3	99.2	0.8
20	Diatrizoic acid	2	99.4	99.6	**99.5**	**0.5**
21	Diclofenac	8	92.3	98.1	**95.9**	**4.1**
22	Diglyme	2	62.5	64.2	63.4	36.6
23	Dimethenamid	4	99.2	99.7	**99.5**	**0.5**
24	Dimethomorph	2	99.2	99.5	**99.4**	**0.6**
25	Diuron	4	77.6	93.4	87.1	12.9
26	DNOC	2	97.6	99.7	99.2	0.8
27	Erythromycin	2	98.8	99.0	98.9	1.1
28	Estrone	2	97.2	99.4	98.8	1.2
29	ETBE	2	96.4	96.5	96.5	3.5
30	Furosemide	2	98.5	99.2	**98.9**	**1.1**
31	Gabapentin	2	98.5	98.5	98.5	1.5
32	Guanylurea	4	16.3	26.1	20.75	79.25
33	HFPO-DA	2	98.6	99.2	**98.9**	**1.1**
34	HMMM	2	93.2	93.8	**93.5**	**6.5**
35	Hydrochlorothiazide	2	92.8	96.5	95.0	5.0
36	Ibuprofen	2	99.1	99.2	99.1	0.9
37	Iopamidol	2	97.9	98.3	98.1	1.9
38	Isoproturon	4	95.7	99.0	97.9	2.1
39	MCPA	2	99.3	99.3	99.3	0.7
40	MCPP	2	99.3	99.4	99.4	0.6
41	Melamine	2	55.3	70.0	62.9	37.1
42	Metamitron	2	80.3	81.5	80.9	19.1
43	Metformin	6	74.5	87.3	83.1	16.9
44	Methenamine	2	96.4	99.1	**98.2**	**1.8**
45	Metoprolol	6	92.7	97.4	95.5	4.5
46	Metribuzin	2	98.9	99.0	98.9	1.1
47	MTBE	2	95.4	95.4	95.4	4.6
48	Naphthalene-1,3,6-trisulfonic acid	2	99.6	99.7	99.7	0.3
49	Naphthalene-1,5-disulfonic acid	2	99.5	99.6	99.6	0.4
50	NDMA	2	4.3	6.5	5.3	94.7
51	Nicosulfuron	4	99.6	99.8	99.7	0.3
52	NMOR	2	60.0	61.0	60.5	39.5
53	NTA	2	99.8	99.8	99.8	0.2
54	Pentoxifylline	4	94.7	99.3	98.4	1.6
55	PFBA	4	99.1	99.9	99.8	0.2
56	PFBS	4	98.8	99.5	99.2	0.8
57	PFHA	4	99.3	99.5	99.5	0.5
58	PFHS	4	99.3	99.5	99.4	0.6
59	PFNA	4	97.9	99.7	99.1	0.9
60	PFOA	6	99.3	99.6	99.5	0.5
61	PFOS	6	99.5	99.6	99.5	0.5
62	Phenobarbital	2	96.0	97.3	**96.7**	**3.3**
63	Pirimicarb	2	99.5	99.7	99.6	0.4
64	Propranolol	2	86.2	93.3	90.3	9.7
65	Pyrazole	2	8.8	37.3	19.3	80.7
66	S-Metolachlor	4	99.7	99.8	99.8	0.2
67	Sotalol	6	94.7	98.1	96.6	3.4
68	Sucralose	2	91.9	94.6	**93.4**	**6.6**

(*continued on next page*)

Table 3 (*continued*)

Num.	Compound	n	Min	Max	Average removal %	Passage %
69	Sulfamethoxazole	2	99.2	99.3	99.2	0.8
70	TBA	2	14.5	20.9	17.5	82.5
71	TCEP	2	98.1	98.5	98.3	1.7
72	Terbuthylazine	2	98.5	99.0	**98.8**	**1.2**
73	Tetraglyme	2	85.2	88.6	87.0	13.0
74	TFA	2	97.3	98.2	97.8	2.2
75	Tiamulin	2	99.4	99.7	**99.6**	**0.4**
76	TPPO	2	99.2	99.3	**99.2**	**0.8**
77	Tramadol	2	97.0	98.2	97.7	2.3
78	Triglyme	2	80.6	83.2	81.9	18.1

DEP: diethyl phthalate; DNOC: dinitro-ortho-cresol; ETBE: ethyl tert-butyl ether; HFPO-DA: perfluoro-2-methyl-3-oxahexanoic acid; HMMM: hexa(methoxymethyl)melamine; MCPA: 2-methyl-4-chlorophenoxyacetic acid; MCPP: meta-chlorophenylpiperazine; MTBE: methyl tert-butyl ether; NDMA: *N*-nitrosodimethylamine; NMOR: N-nitrosomorpholine; NTA: nitrilotriacetic acid; PFBA: perfluorobutanoic acid; PFBS: perfluorobutanesulfonic acid PFHA: perfluorohexanoic acid; PFHS: perfluorohexanesulfonic acid; PFNA: perfluorononanoic acid; PFOA: perfluorooctanoic acid; PFOS: perfluorooctanesulfonic acid; TBA: T-butyl alcohol; TPPO: triphenylphosphine oxide; TFA: trifluoroacetic acid; TCEP: tris (2-chloroethyl) phosphate.

of log D values, which were not different from log K_{ow} values in this assessment. For most of the compounds pKa and log D were calculated with Chemaxon software [47] and for those for which values were not available in Chemaxon, the ChemSpider database [48] was used. It should be mentioned that the experimental values of selected properties were used when available from literature. MV of compounds was calculated as the molar mass (g/mol) divided by the mass density (g/cm^3). The MV values and polarizability of compounds were extracted from CompTox Chemicals Dashboard [49]. Molecular size is defined as length, width, and depth. The distance between the two most distance atoms were determined as a molecular length [18] which was measured by the commercial software Chem3D Ultra 7 [50], and the molecular width and molecular depth which width > depth [12] were calculated by the HyperChem software [51]. The eqwidth was defined as the geometric mean of width and depth [12]. The water solubility of MPs was extracted from the ChemSpider and PubChem database.

2.6. Assessment of solute removal

The solute removal by membrane can be described by both rejection and passage phenomenon. Eq. (1) shows the solute (%) which has been removed by RO membranes. The passage (Eq. (2)) describes the quantity of solute, as a percentage, which passes through the RO membrane into the permeate stream. For better clarification both passage and rejection of solutes were assessed in this study. Rejection and passage were calculated using Eqs. (1) and (2), respectively.

$$R\ (\%) = \left(1 - \frac{C_{ROP}}{C_{ROF}}\right) \times 100 \tag{1}$$

$$P\ (\%) = \left(\frac{C_{ROP}}{C_{ROF}}\right) \times 100 \tag{2}$$

where C_{ROP} is the permeate concentration (µg/L) and C_{ROF} is the bulk feed concentration (µg/L).

Moreover, for compounds with concentrations below the detection limit, the removal percentage was calculated with respect to the half-value of the detection limit [52] which was indicated in the results (Table 3) in bold. Moreover, since the MPs concentration was measured in several replicates, to determine the average MPs removal, the specific MP removal percentages were transformed to a logit-scale through Eq. (3) in order to obtain normal distributed data and the average of the logit transformed removals were determined. Then, this value is transformed back to a percentage removal.
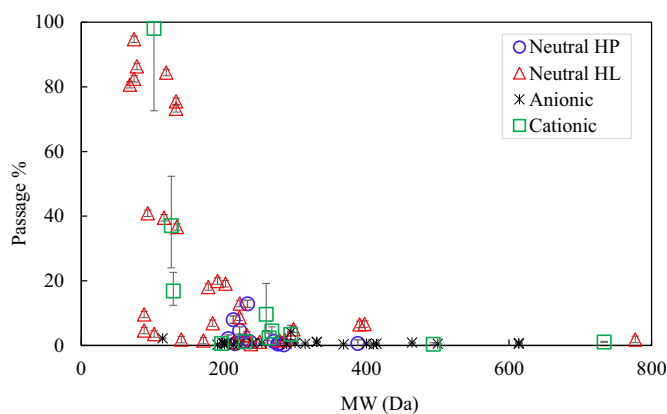

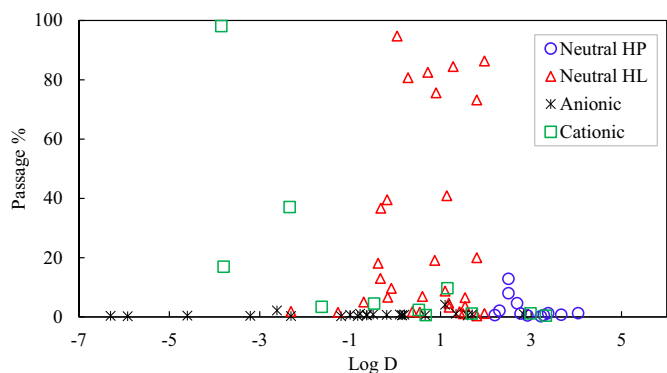**Fig. 1.** Effect of molecular weight on MPs passage by the ESPA3 RO membrane.

Fig. 2. Effect of log D on MPs passage by the ESPA3 RO membrane.

$$Logit(R) = ln\left(\frac{R}{1-R}\right) \tag{3}$$

where R is the value of MPs removal.

2.7. Statistical analysis

One-way ANOVA was applied to test the hypothesis that the four groups of physical-chemical properties compounds had the same Log-passage means. The Tukey Honestly Significant Difference (HSD) test was used for conducting post-hoc tests on a one-way ANOVA. The ANOVA was applied to the categories 1, 2, 3 and 4 explained in Section 2.5. The Spearman rank correlation coefficient (r) was applied to assess the correlation between physical-chemical properties and removal of all MPs.

Factor Analysis was applied through a preliminary PCA which was reduced the contribution of the less significant variables [53]. For rotating the PCA axes, the Varimax rotation criterion was used which turning PCA into a Factor Analysis (FA) [53]. The preliminary analysis was carried out with PCA to examine the contribution of selected physical-chemical properties in each component.

Multiple Linear Regression (MLR) analysis was applied to predict the compound's removal. A stepwise procedure which was used in many environmental assessments was utilized where each variable is entered in sequence [12,54,55]. The variation inflation factors (VIF) of the independent variables were also tested. The Durbin-Watson test was used to examine the residual autocorrelation. Finally, in order to investigate the impact of feed water temperature on the MPs' removal, a paired *t*-test was applied to the removals of the same compound in two different temperature conditions.
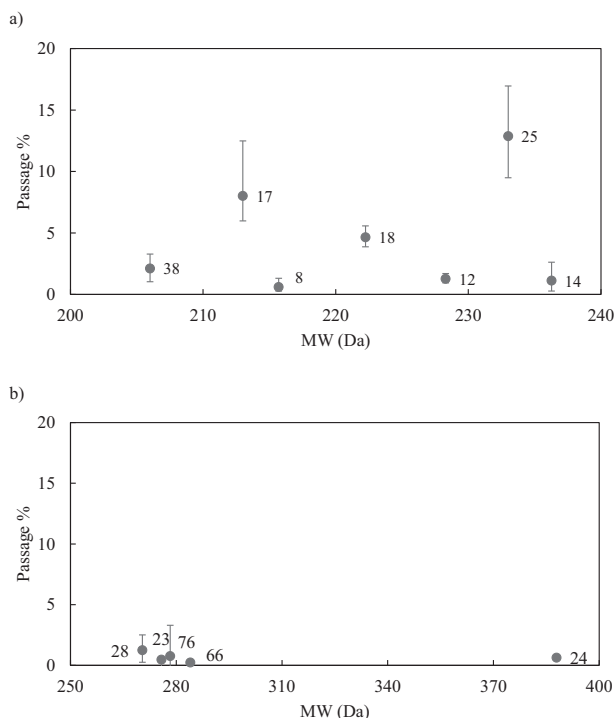
a)

b)

Fig. 3. Effect of MW a) between 200 and 240 Da and b) between 250 and 400 Da on the passage of neutral HP compounds.

3. Results and discussion

3.1. Removal and passage of compounds

The removal and passage of compounds were calculated, and the results are shown in Table 3. In general, passage of the selected compounds decreased with increasing their MW (Fig. 1). However, some compounds with MW lower than 140 Da showed variations in passage. Also, MPs hydrophobicity might be an additional factor to passage variability. However, Fig. 2 shows that hydrophobicity (expressed as log D) alone was not correlated with compound passage indicating that size exclusion was more dominant than the hydrophobicity in removal mechanism of MPs in this study.

3.2. Removal of neutral HP MPs (category 1)

The size exclusion and adsorption of HP compounds onto membranes were recognized as important factors in the removal of neutral HP MPs [22,27]. In our study, the neutral HP MPs were considerably removed. All selected compounds indicated a passage lower than 5% (Fig. 3) with the exception of diuron (25) and chlortoluron (17). Although size exclusion demonstrates the main factor in removal of neutral MPs, the influence of solute-membrane affinity interactions in removal of HP MPs should not be ignored. In fact, neutral HP MPs can adsorb to the membrane because of their log D which >2 and the affinity between hydrophobic moieties of the compounds and the active layer of membranes [26,56,57].

The spearman correlation (Tables S5 and S6 in Supplementary material Section 2.2) revealed that the passage of neutral HP MPs was significantly correlated with their MW (r = −0.63; p-value = 0.02). However, this correlation was weaker than the neutral HL MPs (r = −0.67; p-value <0.001). This could be in regard to the combined effect of size exclusion and adsorption in removal mechanism of neutral HP compounds [56,57].

Regarding to the feed water matrix effect on HP removal, Majewska-Nowak et al. [58] found that atrazine (8) with passage of 6% could
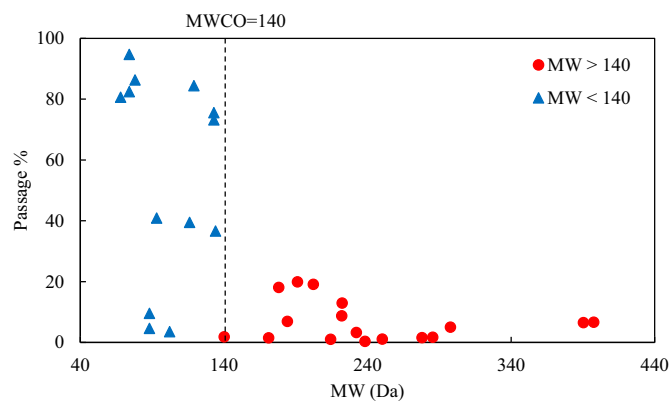

Fig. 4. Effect of MW and MWCO on the passage of neutral HL compounds.
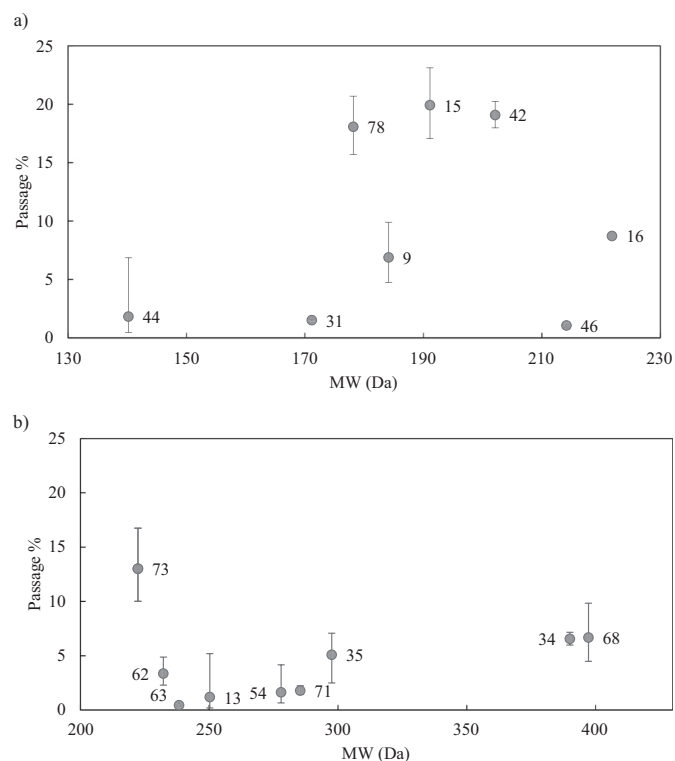

a)

b)

Fig. 5. Passage of neutral HL MPs with MW higher than MWCO (140 Da); a) MW between 130 and 220 Da; b) MW between 220 and 400 Da.

adsorb to organic matter of the feed water which increased its size. Thus, the removal of atrazine will be increased as a consequence of size exclusion and electrostatic interaction between the membrane surface and organic matters. As a result, the hydrophobic–hydrophobic interactions between solute and membrane with the contribution of steric hindrance were the most significant mechanisms for the removal of HP compounds. The removal of diuron (25) and chlortoluron (17) were lower than other compounds in this group and the passage of them through the RO membrane were 12.9% and 8% respectively. Fujioka et al. [59] indicates that size exclusion is not the only mechanism for removal of diuron and chlortoluron by RO. Since diuron and chlortoluron do not have an atom with a high partial negative charge, the cooperation of hydrogen bonding with amide functional groups of membrane was responsible for the low removal of these compounds [59].
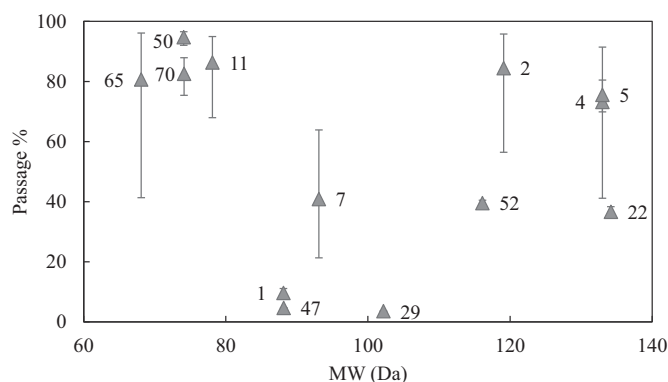
Fig. 6. Passage of neutral HL MPs with MW lower than MWCO (140 Da).

3.3. Removal of neutral HL MPs (category 2)

A sieving effect is the main mechanism for the passage of neutral HL compounds through the RO membranes meaning that the larger the molecule the higher its removal [23,32,60,61]. Besides, since the HL compounds are soluble in water, the coalition of the HL molecule with water molecules might cause a bigger effective diameter of the molecule that consequently results in a higher removal. The MW distribution of uncharged HL MPs ranged between 68 Da and 400 Da, with Iopamidol being the only outlier (777.09 Da).

The overall removal of some MPs was very high supporting the hypothesis of the sieving mechanism. For compounds with higher MW, the passage was lower (Fig. 4). However, some compounds were not following this pattern (e.g., 1,4 dioxane, MTBE and ETBE). As we explained before, molecular weight cut-off (MWCO) is one of the main factors influencing the removal of MPs. The cut-off value of ESPA membranes is around 140 Da which was found in other studies [12,62,63]. For better understanding the passage percentage among neutral HL MPs a cut point of 140 Da was considered for the category 2 compounds and the results are shown in Figs. 5 and 6.

According to Fig. 5, all neutral HL MPs larger than 140 Da were highly or moderately removed (passage < 20%). For some compounds such as methenamine (44), gabapentin (31), metribuzin (46), pirimicarb (63), bisphenol S (13), phenobarbital (62), TCEP (71), pentoxifylline (54) and hydrochlorothiazide (35) the removal was high, and the passage was lower than 5%. This could be a result of steric and electrostatic exclusion. For compounds that are moderately removed (passage higher than 5%) Bellona et al. [32] argued that more than MW, other factors may determine the percentage of hydrophilic removal such as the relationship between the pH of feed water and pKa; and the relationship of molecular width (MWd) and membrane pore size.

For hydrophilic compounds (log D < 2) when the pKa is higher than the feed water pH (pKa > pH), the MWd should be considered. The hydrophilic compounds, having a MWd greater than the membrane pore size (MWd > pore size), are highly to moderately removed. In contrast, when the MWd is smaller than the membrane pore size, low to moderate removal will be observed. Knowing that the feed water pH in our study was around 7 and the pore size of the RO membrane is 0.2 nm (Table 2), we can say that sucralose (68; pKa =11.9; MWd = 0.7 nm), barbital (9; pKa =8.48; MWd = 0.56 nm) and carbendazim (15; pKa = 9.7; MWd = 0.52 nm) have followed the explained pattern showing moderate removals.

Fig. 6 shows the passage of neutral HL compounds with size lower than the MWCO. According to the scientific literature [23,32,60,61], the removal of MPs by RO was highly correlated with their MW (Fig. 1). Thus, for the most of small MPs, the removal was low. However, there are some exceptions (e.g., 1,4 dioxane, MTBE and ETBE) that should be considered.

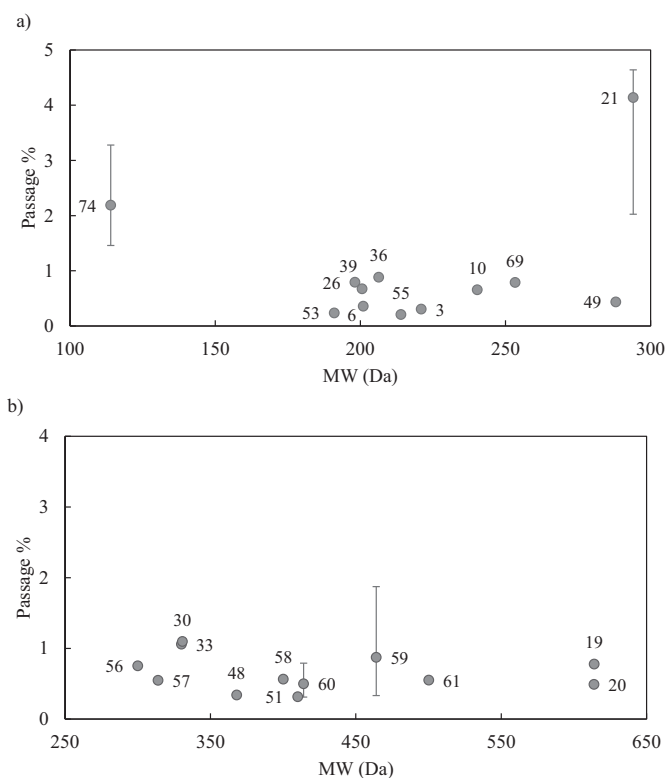1,4 Dioxane (1), MTBE (47) and ETBE (29) are categorized as small

Fig. 7. Effect of MW a) between 100 and 300 Da and b) between 300 and 650 Da on the passage of anionic MPs.

MPs which were almost completely removed (>90%), compared to the others. The explanation for the high removal percentage at a low molecular weight is that 1,4-dioxane, MTBE and ETBE are much more hydrophilic and more polar than others e.g., benzene (78 Da). Benzene (11) thereby adsorbs and permeates through the membrane, while for example, 1,4 dioxane does not [64]. According to some literature, benzene is classified as a hydrophobic solute [65]. However, in the database that was used in this study for measuring the hydrophobicity of compounds, benzene's log D was 1.97 which is very close to 2. Schoonenberg Kegel et al. [65] speculated that the high passage of benzene was probably due to the partitioning of this solute into the membranes as a result of hydrophobic-hydrophobic interactions between benzene and membrane, resulting in an increased transport through the membranes.

NDMA (74 Da) showed 94.72% passage suggesting a very poor removal efficiency. This could be due to the fact that the breakthrough of NDMA is rapid and the removal instantly reaches a constant value [35]. Thus, NDMA was not absorbed by the membrane matrix. According to the studies [66,35] which compared the role of hydrophobicity and the molecular size on MPs removal, the non-adsorbing compounds were removed due to size exclusion, whereas the removal of compounds that adsorbed to the membrane were influenced by hydrophobicity. Therefore, the poor removal of NDMA is more related to its size rather than its hydrophobicity. Moreover, the very compact structure and hydrogen bonding of NDMA might be other reasons of very poor removal efficiency [59,65].

3.4. Removal of anionic MPs (category 3)

The low-pressure ESPA3 RO membrane displayed a net negative charge at operating feed pH (isoelectric point at pH 4 and zeta potential of −24.8 mV at pH 9) [67,68]. The repulsion between the negative charge of the membrane and negatively charged solutes decrease the absorption and increase the removal [23,32]. As a result, the anionic
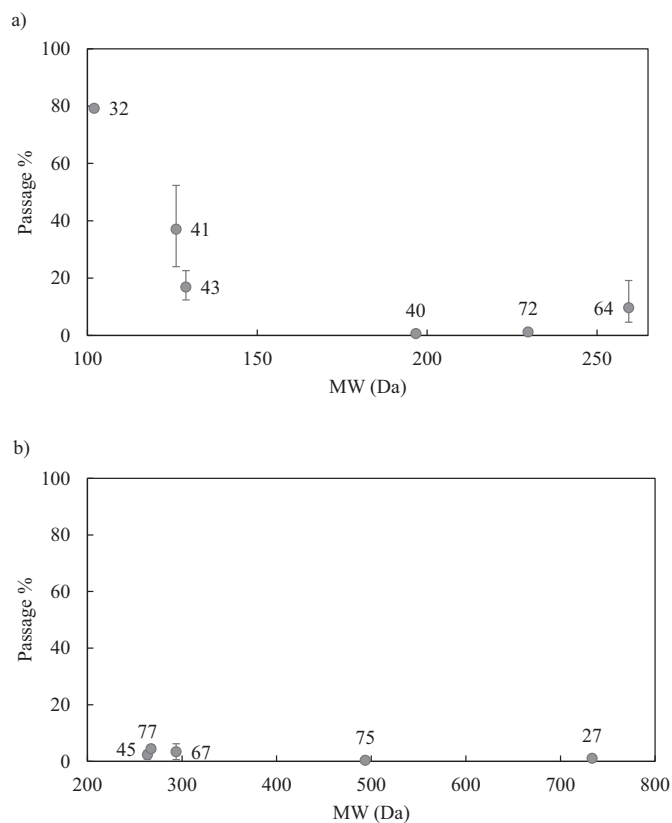
a)

b)

Fig. 8. Effect of MW a) between 100 and 250 Da and b) between 250 and 750 Da on the passage of cationic MPs.
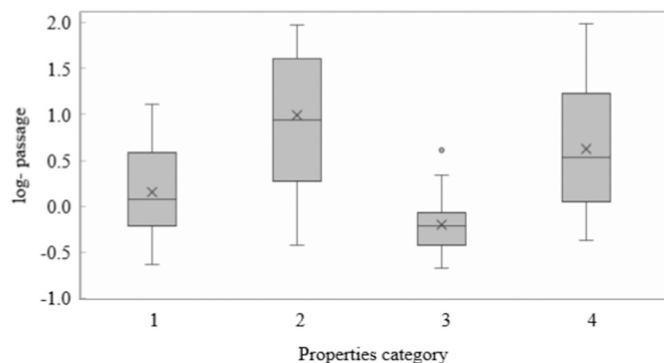

Fig. 9. Box-and-Whisker plot of log-passage range for property categories; 1: neutral HP, 2: neutral HL, 3: anionic, 4: cationic.

compounds were removed very well and passage values lower than 1.5% were observed in most cases (Fig. 7). Also, due to the charge repulsion between anionic compounds and the membrane, the low MW compounds (e.g., TFA) were also highly rejected by the membrane [69].

3.5. Removal of cationic MPs (category 4)

The removal of cationic compounds which was expressed as passage through the RO membrane is shown in Fig. 8. The results indicated an acceptable removal (>95%) of cationic MPs by RO and for most of the compounds the passage was <5% with the exception of guanylurea (32), melamine (41), metformin (43) and propranolol (64). However, the electrostatic attractions between cationic MPs (positively charged) and negatively charged active layer of membrane influence the charge concentration polarization, leading to lower removal [25, 44,70].

Moreover, the inverse relationship between MW and passage (Fig. 8) displayed the effect of size exclusion in removal of cationic MPs.

Metformin (129 Da) was one of the small cationic compounds considered in this study, and it showed lower passage (16.9%) than another small neutral compound 1H-benzotriazole (119.1 Da) with passage of 84.4%. This comparison illustrated that for small cationic MPs, the additional electrostatic interactions between compounds and membrane may influence their diffuse through the membrane which negatively charged resulting in a lower passage. As a conclusion, for removal of cationic MPs, size exclusion, electrostatic sorption [25,44,71] and Donnan exclusion [72] were considered as dominant factors.

3.6. Statistical analysis

The box and whisker plot was generated based on log-transformed of MPs passage values (Fig. 9). The result highlights the lower passage of category 3 (anionic MPs) compared to categories 1 and 2 (neutral MPs) and category 4 (cationic MPs) compounds.

3.6.1. Preliminary analysis

The one-way ANOVA test was run based on these four categories. The result indicated that there was a statistically significant difference between four property categories in terms of their passage through the membrane (p-value < 0.00004). The Tukey's multiple comparison test showed a significant difference between category 1 and 2 (p = 0.00035), between category 2 and 3 (p = 0.000002) and between category 3 and 4 (p = 0.001); whereas the mean passage difference between category 1 and 3 (p = 0.23), category 1 and 4 (p = 0.23) and category 2 and 4 (p = 0.26) were found not significant. Table S3 in Supplementary materials (Section 2.1) shows the result of Tukey's HSD test with the significance level among four categories.

These results illustrated that there was a significant relationship between MPs passage and charge and hydrophobicity of compounds. For anionic compounds (category 3), the impact of electrostatic repulsion in passage of MPs through the membrane was highlighted. Another interesting result was the effect of hydrophobicity which influenced significantly on the passage of neutral MPs in our study. Spearman correlation analysis (Table S4, Supplementary materials Section 2.2) was applied to the data of all MPs (78 compounds) and the result indicated that passage and MW had a moderate inverse correlation (r = −0.59; p < 0.001), showing the overall relationship between the compound size and their removal. Tables S5–S8 (Supplementary materials Section 2.2) show the Spearman correlation between passage and physical-chemical properties of MPs among four categories.

Our results indicated that in RO process, the transport of MPs is mainly governed by the interactions between size, charge, and hydrophobicity, while other physical-chemical properties (e.g., solubility, molecular geometry, polarizability) may influence the compounds removal. Therefore, further evaluation was conducted by means of PCA with a particular focus on these other compound's properties.

3.6.2. Principal component analysis (PCA)

PCA was applied to eleven physical-chemical properties (Table 1) in order to reduce the physical-chemical property sets that would significantly represent a compound and constitute the future predictors for MLR analysis. These variables were molecular weight (MW), molar volume (MV), solubility, polarizability, log K_{ow}, log D, pKa, molecular length (length), molecular width (width), molecular depth (depth) and equivalent molecular width (eqwidth). The analysis of variables by PCA before MLR may in fact help to identify variables that show a co-variability and avoid the multicollinearity problem of MLR models. After applying the PCA, three principal components were extracted, accounting for 73.78% of total variance.

Rotated component 1 (F1: 40.62% of total variance) was related to the size of the molecules (MW, MV, length, width, depth, eqwidth), and
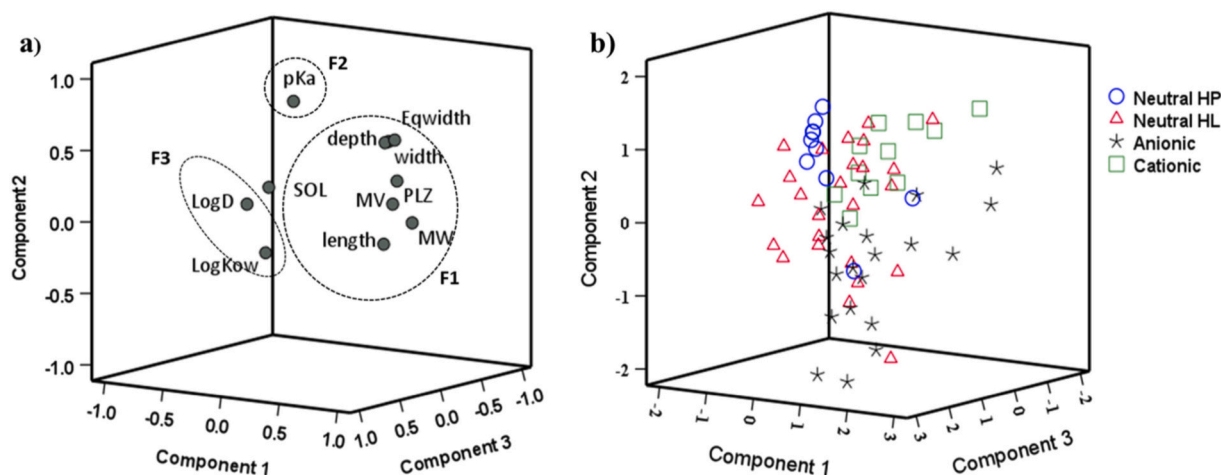
Fig. 10. Component plot in rotated space (a) and score plot (b) of rotated principal components (HP = hydrophobic; HL = hydrophilic; PLZ = polarizability; SOL = solubility).

Table 4
Multiple Linear Regression model to predict the removal of selected MPs.

R: 0.80	F: 28.43	p-Value < 0.001		Durbin-Watson: 2.00	
R^2: 0.68					
	B (CI 95%)	β	t	p-Value	VIF
(Constant)	87.496	–	48.48	0.000	–
Eqwidth	23.834	0.90	6.98	0.000	3.03
Solubility	−10.905	−0.46	−5.81	0.000	1.17
pKa	−7.392	−0.30	−3.86	0.000	1.11
width	−12.334	−0.49	−3.53	0.001	3.46

to their polarizability and it was also inversely correlated with solubility of compounds. Rotated component 2 (F2: 17.17% of total variance) was correlated to the acid/basic strength in solutes (pKa) and rotated component 3 (F3: 15.98% of total variance) was related to hydrophobicity (log K_{ow} and log D). Fig. 10a shows the loading plot of three rotated components revealing the clustering of variables that represents rotating component 1 (F1; size, solubility, polarizability), rotated component 2 (F2; pKa) and rotated component 3 (F3; hydrophobicity). This plot demonstrates clustering of variables which represent the components. Scores of the rotated principal components are presented in Fig. 10b. This plot helps to visualize the removal patterns of 78 compounds in terms of their property categories (three factor loadings). Fig. 10b shows that four compound groups are distinguished in the graph and neutral HP and cationic compounds clustering are observed clearly meaning that size exclusion and hydrophobicity were the mechanisms of removal for these groups. However, not all cases cluster due to the influence of size exclusion or hydrophobicity of the components. For the removal of some compounds (e.g., anionic), the membrane properties also played a role. Therefore, the most appropriate sets of variables were evaluated and MLR analysis was applied.

3.6.3. Multiple Linear Regression (MLR)

According to the previous results, the mechanism of removal and solute passage were affected by the physical-chemical properties and highly depending on charge, size and hydrophobicity of the compounds. The general QSAR linear equation for removal was prepared in order to get the general idea about the factors influencing the MPs removal (all groups together) by RO. The result of the MLR analysis is described in Table 4, and Fig. S1 in Supplementary materials displays the corresponding scatterplot. The MLR model resulted in an R of 0.80, and an adjusted R^2 of 0.68, mentioned that the model explained 68% of the variance of the removal. The solubility, pKa and molecular width were

inversely correlated with the MPs removal, whereas eqwidth was instead directly correlated. Moreover, the result of the Durbin-Watson test was 2 meaning that the model did not show residual autocorrelation. Also, the collinearity diagnostics result confirmed that there was no multicollinearity in the model (VIFs <3.5). The standardized residuals were plotted against the standardized predicted values in Fig. S2 (Supplementary material Section 2.3) and no patterns were present. Therefore, the QSAR linear equation model for rejection could be as follows:

$$rejection = 23.83\,eqwidth - 12.33\,width - 7.39\,pKa - 10.90\,solubility + 87.49$$

(4)

Consequently, a mechanistic interpretation of MPs removal by the MLR model is that the removal will be increased with increasing equivalent molecular width (eqwidth) because of steric hindrance. However, the contribution of eqwidth in size exclusion was compensated by molecular width in final rejection [18]. The equation also shows that a high solubility will decrease the removal. The acidity constant (pKa) which is related to the feed water pH and charge of the compounds showed that the more the acid compounds the better they will be removed by RO membrane. Also, the MLR result proved that although the MW was an important factor in MPs removal by RO, it might not be accurate enough for describing the solute size due to the impact of geometrical configuration on the compound size. The understanding offered by MLR suggests that to improve the removal prediction it would be beneficial to use the reduced number of predictors. Based on such reduced set of variables, more sophisticated algorithms such as artificial neural network (ANN) models could be developed in order to predict rejections of MPs by RO membranes [13].

3.7. Influence of feed water temperature

The overall removal efficiencies of 26 MPs were investigated at two different feed water temperatures (T = 5 °C and T = 19 °C) in order to determine the effect of feed water temperature on MPs passage through the membrane (Table S9). The paired *t*-test was applied to compare the passage of compounds in the two conditions. The result showed that the passage of MPs at a feedwater temperature of 5 °C was significantly lower than at a temperature of 19 °C (p-value = 0.02) suggesting that the MPs removal decreased when feed water temperature increased.

In the RO solution-diffusion model, transport occurs only by diffusion [73]. Thus, as feed water temperature increased, the ion osmotic pressure will increase, which causes an increased pressure gradient in order to sustain the permeation [74]. Moreover, the diffusion coefficient of a compound itself is temperature dependent [73] which influencing the concentration polarization.
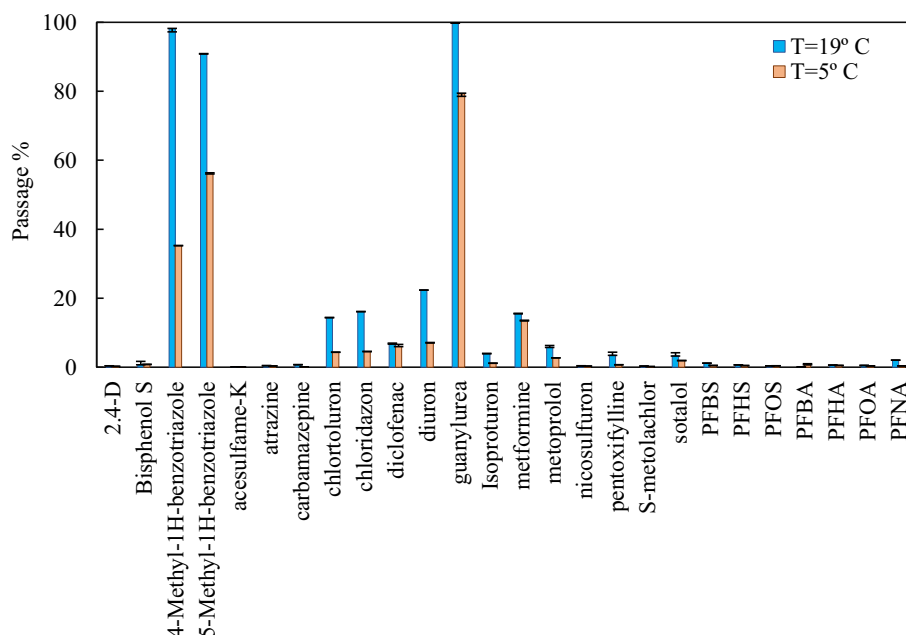
Fig. 11. Comparison between MPs passage in two different feed water temperatures.

On the other hand, higher temperatures increase the viscosity of water, which will more easily diffuse through the membrane [75,76]. Therefore, the flux will be higher which also influences the MPs passage.

In addition, the increase in the feed water temperature caused a rise in the membrane pore size which reduced hindrances to neutral solute [77,78]. Fig. 11 shows the comparison between MPs passage at the two temperatures. Although the higher temperature influences the passage of all compounds, it is obvious that the temperature variation did not have the same effect on all MPs. For instance, the colder temperature had positive effects on 4-Methyl-1H-benzotriazole passage, as the decrease in temperature resulted in 63% additional removal compared to the higher temperature (T = 19 °C). On the other hand, a lower temperature increased the removal of acesulfame-K only by a 0.1%. However, understanding the variations in the removal efficiencies of each MP in the given temperature could be difficult and needs more investigation [79]. Also, the paired t-test results indicated that the differences between removals in two temperatures were more significant for benzotriazoles, chlortoluron, chloridazon, diuron and guanylurea. In any case, it is important to mention that the feed temperature should be kept constant during the treatment.

4. Conclusions

In this study, the removal of 78 MPs by RO filtration was investigated, and four categories were defined for MPs based on their charge (neutral, anionic, cationic) and hydrophobicity (log D). The results proved that RO is a robust barrier for removal of most MPs. 64 MPs showed a high removal with rejection ranging between 80% and 99.8%. Six MPs were rejected moderately from 60% to 80% and eight compounds were poorly rejected (between 2% and 27%). The passage figures for the selected compounds were also created. The varying results were mostly based on MPs characteristics and membrane properties. Neutral and hydrophilic MPs showed the highest passage while anionic MPs displayed the lowest passage through the ESPA3 RO membranes. The statistical analysis revealed that physical-chemical properties of compounds had a significant influence on MPs passage and removal. The size of neutral and hydrophobic MPs was inversely correlated with their passage. However, this correlation was stronger for neutral and hydrophilic MPs. For neutral compounds, the removal was mainly associated with size exclusion. By contrast, the removal of charged MPs was mostly

attributed to charge interactions.

Also, membrane properties (e.g., membrane charge, MWCO and pore size) and feed water properties (e.g., temperature) were determined as key factors. The comparison between MPs passage at different feed water temperatures (5 °C vs 19 °C) showed that the performance of RO membranes may be highly dependent on the temperature of feed water so that the MPs passage through the membrane increases by increasing the temperature. A QSAR model equation could be developed integrating the information about the interaction of solute properties in order to predict the removal of MPs during RO filtration. In addition, the QSAR model identified that compound geometry, charge and solubility were the most important variables that may influence the removal of MPs. Moreover, the result showed that the equivalent width of molecules might be a better predictor than MW for rejection of compounds.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jwpe.2021.102164.

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