

## Application of an integrated process for the removal of organic compounds of the phenols group from water

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### ABSTRACT

We classify bisphenols as organic compounds from the group of phenols. Since bisphenols are carcinogenic and disrupt the human body's hormonal balance, removing these compounds from the aquatic environment is crucial. The production of creams and household plastics uses bisphenols. Such harmful compounds include bisphenol A and its derivatives (e.g., bisphenol B (BPB), bisphenol AP (BPAP), bisphenol M (BPM), bisphenol E (BPE), and bisphenol C (BPC12)). Although bisphenol A is still the dominant bisphenol in the environment, the legal restrictions on its use lead to the use of other bisphenols. Biochemical processes remove 50%–90% of these compounds from wastewater. Introducing Fenton's reagent or UV radiation improves efficiency. However, these are still ineffective methods of their removal from sewage and water. The paper focuses on the possibility of removing bisphenols: bisphenol A, bisphenol E, bisphenol B, bisphenol G, bisphenol C, bisphenol AP, and bisphenol M from surface water using integrated nanofiltration–reverse osmosis membrane processes. The research is carried out at a small nanofiltration and reverse osmosis station. The process uses nanofiltration and the osmotic membrane of spiral aromatic polyamide membranes. Chemical determinations are made by chromatographic analysis developed for these compounds. The conducted studies showed the removal of the tested compounds with high effectiveness. The process parameters significantly influence the removal process, particularly the concentration of bisphenols in the feed and the transmembrane pressure. The impact of these processes should be further analyzed and explained.

*Keywords:* Bisphenols; Water; Nanofiltration; Reverse osmosis; Molecular weight

### 1. Introduction

Bisphenols are hazardous for the human body, disrupting the functioning of the endocrine and reproductive systems, and cause obesity, cardiovascular diseases, breast cancer, and congenital disabilities [1]. The best-known and most commonly used compound in this group is bisphenol

A (BPA). BPA is readily and cheaply synthesized from phenol and acetone, contributing to its wide industrial use [2]. BPA is detected in municipal and industrial wastewater in concentrations up to mg/L and leachate from landfills [3–5]. In contrast, several dozen µg/g are found in sewage sludge [3–5]. Incomplete wastewater treatment, sewage sludge management, and inadequate protection of municipal landfills

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are the primary sources of BPA infiltration into surface and groundwater, where BPA concentrations can reach several dozen  $\mu\text{g/L}$  [4–10]. Biomonitoring studies show that human exposure to BPA is continuous and rapidly developing, which results in its presence in the body of every human being [11–14]. Due to the proven toxic effect on organisms, limitations in its use have been established by the European Commission, the US Environmental Protection Agency, and Health Canada [12–15]. Legal restrictions and increased social concerns have replaced BPA in industrial production with other compounds of similar structure, especially bisphenol analogs, which exhibit the same or improved plasticizer properties. Although the initially alternative BP was considered less toxic than BPA, recent reports show they have similar or even more significant side effects than BPA [4,14–19]. BPs exhibit comparable BPA biological activity, including potential hormonal disruptions, toxicity, and genotoxicity. Bisphenol B (BPB) and bisphenol C show higher estrogenic activity than BPA [3,8,18,20]. The order of toxicity (14 d for EC50) was found to be as follows: bisphenol G > bisphenol X > bisphenol AF > bisphenol A > bisphenol Y > bisphenol M > bisphenol P [21].

Due to their increasing use, BPA analogs have been detected in various environments [22–24]. However, knowledge about their spread and how they enter the environment is still insufficient. Research in this area is very complicated due to: low BP concentrations at the level of  $\text{ng/L}$ – $\mu\text{g/L}$ , the complex composition of the matrix, and the lack of standard analytical methods.

Bisphenols' dangerous biological activity and toxicity necessitate research on their removal in water and wastewater treatment processes. The most effective processes are based on the use of microorganisms. They remove compounds that are difficult to eliminate by conventional water purification methods [25]. The most common method of BP removal is adsorption with appropriate adsorption capacity and thermodynamic parameters of the process [26]. Next are the biodegradation mechanism using native enzymes, membrane separation processes, photocatalytic oxidation, nanosorbents, and thermal degradation [27–29].

Studies of BPA removal from wastewater and water by high-pressure methods have yielded efficiencies of 65%–90% [30–33]. However, the removal of bisphenol A from water and wastewater by membrane methods is still under investigation, as the efficiency of removal of this contaminant is determined by the physicochemical properties of the compound, the organic and inorganic membranes and the parameters of the nanofiltration (NF) and reverse osmosis (RO) process itself [31–33].

Recent literature reports related to bisphenol A removal have focused on the choice of membrane-forming material [33–36] as it significantly impacts the efficiency of the process. Yuksel et al. evaluated the removal of BPA from ultra-pressure water in an integrated NF and RO process by several types of membranes, analysing parameters such as water permeability coefficients, retention factor and permeate flux, from which it was concluded that the efficiency depends on the membrane-forming material. Furthermore, BPA is classified as a hydrophobic inert compound but acidic, affecting the interaction with the membrane surface and the removal efficiency in membrane processes [36–38].

On the other hand, investigating the efficiency of removing bisphenol analogues from water using NF and RO in a single or integrated system requires research, given the recognition of the mechanism of removal efficiency.

The effectiveness of ultrafiltration, nanofiltration and reverse osmosis in the treatment of water and wastewater from bisphenol A was only conducted as single processes. Furthermore, the research focused only on bisphenol A while other bisphenol analogues were completely ignored. The choice of the type of pressure process was mainly determined by the type of contaminants present in the water streams to be treated. Accordingly, nanofiltration and reverse osmosis are the most suitable processes for the removal of small-molecule organic contaminants with a diameter of approximately 1 nm or a molar mass of 150–500 g/mol. This is primarily due to the physico-chemical properties of nanofiltration and reverse osmosis membranes [39].

The scientific literature presents examples documenting micropollutant retention rates in ultrafiltration ranging from 17% to 80% [30]. Micropollutant retention in this case was caused by the adsorption effect of the compound on the membrane.

The research presented in this paper is the treatment of surface water from bisphenol A and its derivatives using an integrated NF-RO process.

## 2. Research methodology

### 2.1. Technological research

The semi-technical pilot plant was designed by Stadtwerke Düsseldorf (SWD) and realized by Cornelsen Umwelttechnologie GmbH, Essen, Germany. The low-pressure reverse osmosis (LPRO) unit was constructed by Grünbeck Wasseraufbereitung GmbH, Höchstädt a.d. Donau, Germany (GENO-Nano RKF1800 S). An integrated system was used for the study: nanofiltration–reverse osmosis. During the experiments, the transport-separation properties of the membranes in both filtration stages (NF and RO) were determined on the basis of the permeate volume flux values and the degree of concentration reduction of the bisphenols tested. The first stage was the removal of bisphenols by nanofiltration, during which the transmembrane pressure and the initial concentration of bisphenols in the surface water were altered. The treated water after nanofiltration was introduced into a reverse osmosis system. Water samples after NF and RO were taken for chemical analysis. A spiral module was used in the test set-up.

The research on the removal of bisphenols was carried out in the NF and RO processes. 60% recovery rate. In both systems, composite membranes (polyamide skin layer) were used.

Since surface water was used for the study, it was therefore necessary to use membrane protection against iron, manganese ions, scaling and fouling, this was done by pretreating the surface water on a sand filter.

After the tests, the membrane was rinsed with deionized water, thus determining the change in transport properties of the membrane after the operation. The effectiveness of the water filtration process was assessed based on the transport and separation properties. A mixture of bisphenols was used in the research:

Table 1  
Parameters of the nanofiltration and reverse osmosis process

	Nanofiltration (KeenSen)	Reverse osmosis (FilmTec)
Active membrane surface	0.3 m <sup>2</sup>	0.28 m <sup>2</sup>
Membrane material	Polyamide	Polyamide
Cut-off	350 Da	150 Da
Concentrate flow rate	5.5 L/h at 50% recovery (conversion)	5.5 L/h at 50% recovery (conversion)
Permeate flow rate	5.5 L/h at 50% recovery (conversion)	5.5 L/h at 50% recovery (conversion)
Capacity/flow rate	75 GPD/284 L/d	75 GPD/284 L/d
Transmembrane pressure	1.15 MPa	1.5 MPa

Table 2  
Process parameters with the use of reverse osmosis and nanofiltration

	Nanofiltration					
Transmembrane pressure (MPa)	1.15	0.9	1.15	0.9	1.15	0.9
Total concentration of bisphenol in the feed (μg/L)	150		300		450	
	Reverse osmosis					
Transmembrane pressure (MPa)	1.5	1.3	1.5	1.3	1.5	1.3
Total concentration of bisphenol in the feed (μg/L)	2.31		8.87		17.92	

- Bisphenol F (4,4'-Methylenediphenol) – BPF
- Bisphenol A (4,4'-Isopropylidenediphenol) – BPA
- Bisphenol C (4,4'-Isopropylidenedi-o-cresol) – BPC
- Bisphenol AP (4,4'-(1-Phenylethylidene)bisphenol) – BPAP
- Bisphenol B (4,4'-sec-Butylidenediphenol) – BPB
- Bisphenol E (4,4'-Ethylidenebisphenol) – BPE
- Bisphenol G (4,4'-Isopropylidenebis(2-isopropylphenol)) – BPG

Bisphenol removal studies in the NF-RO system were conducted according to the parameters in Table 2.

The efficiency of the nanofiltration process was determined thanks to the performance measurements (for deionized water, volumetric permeate flux –  $J_w$  and for the research sample –  $J_p$ ), relative volumetric permeate flux ( $\alpha$ ), and the selectivity ( $R$ ) of membranes.

## 2.2. Analytical methodology

Spectrophotometric, pH-meter, and conductometric determinations were performed according to the applicable standards or based on analytical methodologies commonly recognized and recommended by the scientific literature. The test results presented in the paper are the average of at least three determinations performed simultaneously.

## 2.3. Extraction procedure

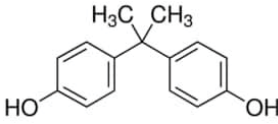
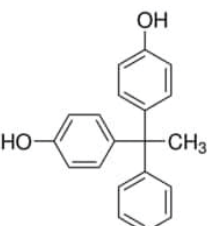
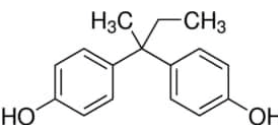
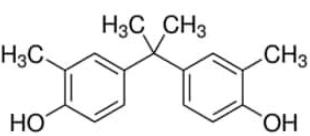
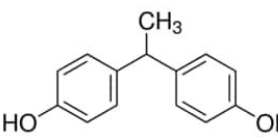
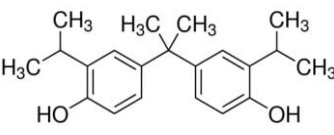
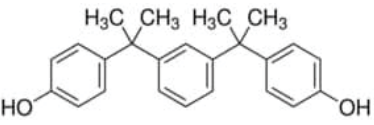
The isolation and derivatization of six bisphenols were realized by ultrasound-assisted emulsification micro-extraction (USAME). 5 mL of water samples and 0.2 g of disodium hydrogen phosphate (4%) were prepared.

The extraction solvent (chlorobenzene, 60 μL) and the derivatization reagent (acetic anhydride, 225 μL) were added to the water sample and mixed. Extractions were performed at 42 kHz of ultrasound frequency and 230 W of power for 5 min at room temperature. Emulsions were disrupted by centrifugation at 4,000 rpm for 5 min in a laboratory centrifuge. In effect, the organic phase settled at the bottom of the conical tube. After centrifugation, carbon tetrachloride was removed using a 100-μL Hamilton syringe (USA) and transferred into a 150-μL microvial with an integrated insert. Gas chromatography–mass spectrometry (GC-MS) analysis was performed as described in GC-MS analysis.

## 2.4. Chromatographic analysis

The current concentration of bisphenols was monitored using an HP 7890B gas chromatograph with an electronic pressure control device connected to a mass detector MSD 5977A (electron impact source and quadrupole analyzer, Agilent Technologies, USA) equipped with the HP-5MS column (5% phenylmethylsiloxane) with the length of 30 m and i.d. of 0.25 mm coated with a 0.25 μm thick film and using a split-less injector at a temperature of 250°C. The helium of 99.999% purity was used at a constant flow rate of 1.0 mL/min as a carrier gas. The electron impact source temperature was 230°C with electron energy of 70 eV. The quadrupole temperature was 150°C, and the interface between GC and MS temperature was 280°C. The oven temperature was programmed from 130°C for 3 min and increased at increments of 30°C/min to 250°C for 4 min and later with the growth of 20°C/min to 310°C for 5 min [29]. The monitored ions were selected based on the analysis of mass spectra recorded by the MS detector in scan mode (Table 3).

Table 3  
Basic information of the six determined bisphenols

Compound	Structure	Molecular mass (g/mol)	$\log K_{ow}^b$	pKa <sup>c</sup>	Henry's constant (at·m <sup>3</sup> /mol)
Bisphenol A		208.29	3.64	10.29	$4.0 \times 10^{-11}$
Bisphenol AP		290.36	4.33	10.22	$5.7 \times 10^{-10}$
Bisphenol B		242.31	4.15	10.27	$1.2 \times 10^{-11}$
Bisphenol C		281.10	–	9.90	–
Bisphenol E		214.26	3.23	10.10	–
Bisphenol G		312.45	6.55	–	–
Bisphenol M		346.46	–	–	–

<sup>b</sup> $\log K_{ow}$  – octanol/water partition coefficient;  
<sup>c</sup>pK<sub>a</sub> – acid dissociation constant.

### 3. Results and discussion

The sand bed filtration process significantly improved the quality of surface water (Table 4) and, at the same time, protected the nanofiltration membrane against fouling and scaling.

Bisphenols were removed from water in the NF process with a high retention rate (Table 5). It was observed that at 150 µg/L bisphenols in the filtrate, all BPs removed significantly less compared to other concentrations (300 and 450 µg/L) regardless of transmembrane pressure. BPs with a higher  $\log K_{ow}$  value have a higher retention factor.

This demonstrates a two-step separation mechanism in nanofiltration – in the first step, the compound adsorbs on the membrane surface, and in the second step, it passes through the membrane by diffusion and/or convection determined by its affinity for the membrane polymer or its ability to dissolve. Of the BPs tested, BPE had the lowest removal relative to the other BPs reaching between 71% and 99%. This is probably due to the lowest value of  $\log K_{ow}$ , which may indicate the dual nature of this compound, that is,  $\log K_{ow}$  qualifies it as a hydrophobic compound while at the same time being highly soluble in water. In addition, certain mechanisms contribute to the removal of this

compound (e.g., exclusion of repulsion size) with adsorption on a membrane. Adsorption, on the other hand, may adversely affect the retention factor, as some compounds may dissolve in the active layers of the membrane and then diffuse through the polymer and eventually desorb on the permeate side [39–42]. This analogue was found to have a higher concentration in the permeate in the last sample of the process suggesting that it probably dissolved in the membrane and then desorbed on the permeate side [39–42]. It is important to point out that the separation of micropollutants is affected by a number of membrane phenomena, including the reduction of the zeta potential and the presence of a filter cake on the membrane surface [43–45]. As shown in papers [4–7,45–48], membrane fouling increases the intensity of the micropollutant adsorption effect and limits the process of compound diffusion through the membrane.

The removal efficiency of bisphenols for the nanofiltration membrane used was influenced by the molecular weight cut-off value (Table 3), which was higher than that of the hydrophobic compounds removed. The cut-off (separation)

layer of the nanofiltration membrane is a negative charge carrier, which minimised the adsorption of negatively charged compounds causing coating of the membrane surface, leading to reduced performance (fouling). This phenomenon is often caused by natural organic substances present in surface waters.

When examining the effectiveness of RO, it was found (Table 6) that the transmembrane pressure from 1.3 to 1.5 MPa had a slight effect on the concentration of the tested compounds in the permeate. In all tested compounds, the treatment efficiency ranged from 92% to 100%.

Regardless of the pressure applied, the weakest removal of BPE was at an initial concentration of 150 µg/L yielding between approximately 94% and 97%. For the other two concentrations (300 and 450 µg/L), BPE retention rates were between 92% and 98%. BPE is removed much more weakly at higher concentrations in the feed.

The lower values of the retention coefficient were at the concentration of 300 and 450 µg/L; the values ranged from 97% to 100%. The tested BPE bisphenols are hydrophobic (determined by the of  $\log K_{ow}$  value Table 3) these characteristics probably affected the interaction with the membrane surface, leading to an electrostatic attraction or repulsion phenomenon.

Similar results were presented by Baransi-Karkaby et al. [41]. In both cases, the quantitative and qualitative composition of the feed played a significant role. The tested bisphenols were BPA, BPB, and BPM. For these three compounds, the retention coefficient at the concentration of 200 µg/L was 95%, increasing the amount of bisphenols, and the coefficient decreased to 60%. In contrast, Baransi-Karkaby et al. [41] and Khazaali & Kargari [42] investigated the effects of various parameters such as pressure, flow rate, feed concentration, and pH on BP removal. The results showed a maximum removal rate of 87.34% for a concentration of 50 mg/L at a pressure of 408.1 kPa, pH 8, and a flow of 1.172 L/min. The most effective influencing parameter was the feed flow rate, which showed an intense concentration polarization at the membrane surface. The effect of feed pH showed minimal effect at pH 10 [42,43].

Table 4

Water used for the tests was (surface water with added compounds from the bisphenol group

Parameters	Surface water			R (%)
	Values		Standard deviation	
	Mean	Median		
Color, mg·Pt/L	6.9	12.0	1.77	97.3
Turbidity, NTU	1.20	1.30	0.79	78
Conductivity, µS/cm	331	388	21.7	52.4
pH	7.15	7.26	0.11	
Calcium, mg·Ca <sup>2+</sup> /L	80.03	84.1	2.70	30.1
Manganese, mg·Mn/L	0.02	0.02	0.05	72.8
Ferrum, mg·Fe/L	0.09	0.38	0.16	87.1
COD <sub>Mn</sub> , mg·O <sub>2</sub> /L	10.5	10.4	1.96	42.5

Table 5

Retention factor (R) of removing bisphenols from water by nanofiltration

Sample number	Total concentration of bisphenol in the feed (µg/L)	Transmembrane pressure (MPa)	R (%)						
			BPA	BPAP	BPB	BPC	BPE	BPG	BPM
1	150	1.15	96.91	99.35	99.51	99.26	98.50	99.28	99.30
2			97.72	99.60	99.62	99.20	99.58	99.30	99.13
1		0.9	92.04	99.18	99.04	99.26	89.02	99.13	99.21
2			96.54	98.72	98.99	99.26	71.90	99.27	98.90
1	300	1.15	99.77	99.97	99.96	99.97	99.66	99.94	99.95
2			99.81	99.95	99.95	99.96	99.71	99.92	99.91
1		0.9	99.64	99.95	99.93	99.94	98.92	99.93	99.86
2			98.94	99.94	99.87	99.93	97.20	99.94	99.95
1	450	1.15	98.91	99.88	99.84	99.94	96.48	99.91	99.95
2			97.79	99.93	99.89	99.96	93.48	99.81	99.96
1		0.9	98.05	99.98	99.89	100.00	94.21	99.94	100.00
2			90.15	99.94	99.45	99.98	91.91	99.98	100.00

Table 6  
Retention factor of removal of bisphenols from water using reverse osmosis water

Sample number	Total concentration of bisphenol in the feed ( $\mu\text{g/L}$ )	Transmembrane pressure (MPa)	R (%)						
			BPA	BPAP	BPB	BPC	BPE	BPG	BPM
1	2.31	1.5	96.07	97.93	97.41	97.65	96.33	97.35	93.10
2			96.73	98.61	97.76	98.65	96.86	98.40	97.13
1	2.31	1.3	96.23	95.10	97.78	97.38	96.04	98.64	94.01
2			95.68	97.74	98.25	98.01	93.57	97.85	92.65
1	8.71	1.5	99.74	99.96	99.89	99.95	98.31	99.96	100.00
2			98.63	99.82	99.84	99.87	94.48	99.91	99.77
1	8.71	1.3	99.63	99.92	99.87	99.90	97.25	99.91	99.71
2			99.65	99.96	99.90	99.93	97.93	99.96	100.00
1	17.92	1.5	98.97	99.50	99.73	99.90	93.04	99.94	100.00
2			99.00	99.90	99.77	99.88	92.78	99.94	99.82
1	17.92	1.3	98.76	99.86	99.68	99.85	92.98	99.94	100.00
2			98.54	99.90	99.56	99.81	92.08	99.93	100.00

Sample number \* – the presented sample is the first and last of the process.

Pre-treated surface water on a sand filter probably influenced the efficiency of removing bisphenols from the water. It is confirmed by [35]. Removal of BPA from ultrapure water on polyamide membranes showed almost complete removal of BPA, pointing out that such high effects can be achieved by pre-treating the water. On the other hand, different results were obtained by [45]. They obtained higher retention of the tested compounds for natural water than for deionized water samples.

It was also found that BPA and its analogs removed with very similar effects in the studied processes. Similar results were observed by [5,33,40], no significant change in BPs removal for polyamide membranes in NF and RO was found.

A common feature of the membranes studied was their epidermal layer made of poly(aromatic amide) [28,43–48], so that both carboxyl and amine groups are present on the membrane surface. In the case of separation of organic micropollutants, the presence of carboxyl groups – which give the membrane a negative charge – enables the formation of so-called hydrogen bridges between the pollutant to be removed and the membrane. The absorption phenomenon is also determined by the presence of polar groups in the micropollutant molecules.

Common phenomena accompanying membrane filtration are fouling and scaling of membranes, which, apart from lowering the membrane efficiency, also change its morphology and surface charge, which affects the removal of micropollutants. In the conducted research, the deionized water volume flow ( $J_w$ ) for RO before treatment of water with bisphenols was  $5.5 \times 10^{-5} \text{ m}^3/\text{m}^2\cdot\text{s}$ , and after the process,  $4.89 \times 10^{-5} \text{ m}^3/\text{m}^2\cdot\text{s}$ , while in NF before 42.1 after  $40.9 \times 10^{-5} \text{ m}^3/\text{m}^2\cdot\text{s}$ . Relative volumetric permeate flux was less than 1 ( $\alpha < 1$ ) and amounted to 0.88 for RO and 0.84 for NF. The obtained transport and separation values indicate slight fouling and adsorption of the membrane.

Comparing the separation efficiency (Figs. 1 and 2) for the concentration of 150 and 450  $\mu\text{g/L}$ , an apparent effect of the bisphenol concentration in the feed on the removal efficiency is observed, especially in the reverse osmosis

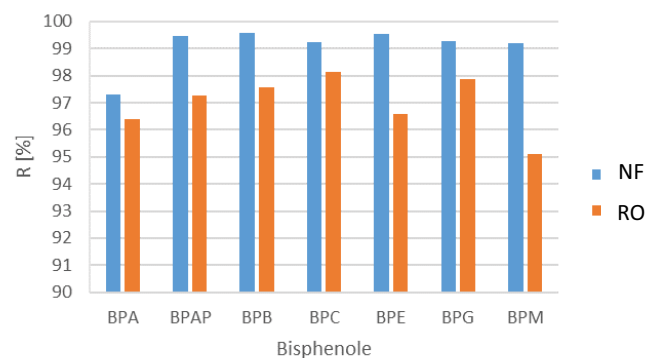


Fig. 1. Comparison of BP removal efficiency in nanofiltration and reverse osmosis 150  $\mu\text{g/L}$ .

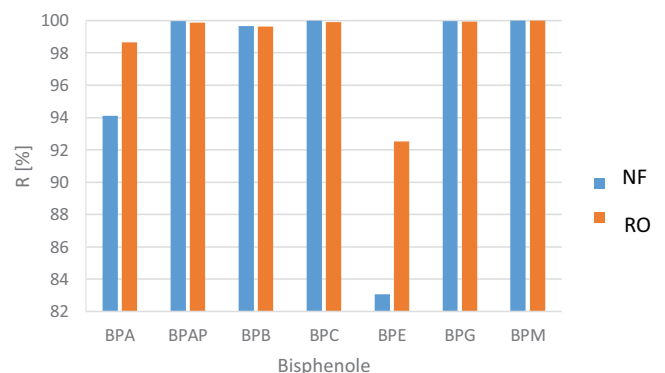


Fig. 2. Comparison of BP removal efficiency in nanofiltration and reverse osmosis at 450  $\mu\text{g/L}$  concentration.

process. Bodzek and Dudziak [44] confirm this observation in their studies, also draw attention to the dependence that the retention coefficient of these processes depends on the molecular weight of pollutants. The higher the mass, the greater the efficiency. No such dependence was observed in our research.

The study showed that the use of two high-pressure processes with different separation mechanisms in an integrated system is an opportunity to achieve drinking water parameters [47].

The mechanism of removal of the tested bisphenols indicates the need for further research because there is no clear answer to the question of what parameters related to the composition of the treated water, the properties of bisphenols and membranes contribute to the effective removal and fouling of membranes and their additional purification or neutralization sequence. Based on the physicochemical characteristics of the bisphenols in question, it can be concluded that the low values of the vapor pressure and Henry's constant indicate the ability of these compounds to accumulate in the aquatic environment as well as their low predisposition to evaporate both from water and soil and that the process self-cleaning is negligible [41,43,44].

In addition, particular attention should also be paid to managing post-regenerative solutions formed after cleaning the membranes, which are likely to be loaded with biologically active organic micropollutants at a higher concentration level than in raw water. It makes it a priority to develop an additional purification or neutralization sequence.

#### 4. Conclusions

Based on the research, the following conclusions were drawn:

- bisphenol removal in both processes removed with a high retention rate of 92%–100%, while nanofiltration alone removes with an average of 98% efficiency, however, concentrations still remain high in the water after the process if drinking water were considered. The proposed solution for the removal of bisphenols from water in an integrated system could be a viable method to improve treatment and affect membrane performance;
- the concentration of feed turned out to be a parameter influencing the process of reverse osmosis in the removal of organic compounds from the bisphenol group;
- to remove bisphenol E, an additional water treatment process should be used due to the low removal retention rate.

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