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Assessment and Removal Strategy of Microplastic Pollution in River Water in the Krueng Aceh River, Indonesia

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ABSTRACT

The pollution of water bodies by microplastic (MP) particles is a significant concern that has drawn the attention of environmental health organizations from various regions of the world. This concern is primarily caused by the potential of particles from the incomplete degradation of plastic waste to enter the food chain via water sources or fish consumed by humans. In Indonesia, the Krueng Aceh River is a water body that stretches across the Aceh Besar Regency and the Municipality of Banda Aceh (Indonesia). The river serves as a raw water source for clean water treatment for residents of both regions. The discovery of MP pollution in rivers in various regions of Indonesia as well as other countries has raised concerns regarding the presence of pollutants in the Krueng Aceh River. Therefore, this study identified MP particle pollution in the Krueng Aceh River water and assessed potential separation using ultrafiltration technology based on Polyethersulfone-graphene oxide membrane. Water samples were collected at five points along the river's flow through the Aceh Besar area and Banda Aceh City. A total of 2 types of flat sheet membranes were created with a composition of Polyethersulfone polymer and graphene oxide in dimethylformamide. The ultrafiltration module was designed using cross-flow filtration with the feed of five samples of Krueng Aceh River water. Analysis was then conducted on the quantity, shape, and type of MP particles in water samples before and after ultrafiltration. The results showed that all water samples contained MP particles at a concentration of 18-22 particles/mL. This indicates that the Krueng Aceh River was already contaminated with MP pollutants. Therefore, special treatment efforts were needed by the government before it could be used as a source for the production of clean water for the residents of Banda Aceh City and Aceh Besar Regency. Based on these findings, the proposed alternative filtration technique can effectively remove pollutants by up to 91%.

1. INTRODUCTION

The world community's concern regarding MP pollution in drinking water has gradually received responses from various health organizations. However, the requirements for drinking water free of microplastics (MPs) have not yet been formulated by governments, the World Health Organization, and the BPOM. The health risks associated with contamination depend on the number of ingested particles and duration of retention in the intestine. Several studies have shown that MPs are plastic components with small sizes (< 5 mm), mostly originating from the breakdown of large-sized plastics [1]. The exponential increase in plastic product demand over the last century has led to approximately 4.8 to 15.11 million metric tons of plastic-based waste entering the ocean annually. Current estimates of the abundance of MPs in the ocean range from 103−105 per m^3 or 0.001−0.1/mL and are not easily removed due to their nonbiodegradable nature [2].

In line with previous studies, MP pollution poses a global threat to marine environments, surface water, sediments, air, soil, food, and drinking water [3]. Laboratory experiments have reported the damaging effects of these materials on marine animals, including behavioral changes, increased mortality, DNA damage, neurotoxicity, and malformations in shellfish [4]. In addition, their accumulation affects humans who consume contaminated water sources or marine animals. In Indonesia, MP pollutants are found in all surface water in the Benoa Bali coastal area and are of various shapes and sizes. MPs of varying sizes were also found in the Surabaya River [5, 6].

The current research on microplastic pollution in rivers worldwide highlights extensive contamination, as demonstrated by studies in various regions. In the Yitong River, China, microplastic pollution is notably higher in urban areas, with most particles being fibrous and ranging in size from 0.05 to 1 mm. The predominant types of microplastics identified are polyethylene, polypropylene, and polystyrene [7]. Significant microplastic contamination was detected in Nigeria's rivers, with the highest levels found in River Benue for fish, Rivers Ogun and Benue for water, and Rivers Yauri and Benue for sediment. The most frequently identified plastics were PET, PP, PE, and PA/Nylon, highlighting the critical need to tackle plastic waste from packaging and textiles [8]. Another study conducted by Maisto et al. [9] revealed significant microplastic pollution in Italy's Volturno River, with concentrations ranging from 1.05 to 14.55 ppm. The most prevalent types were polypropylene (PP) and polyethylene terephthalate (PET), which pose ecological risks at hazard levels III and IV according to the Polymer Hazard Index (PHI). Microplastic pollution has also been documented in several other rivers, including the Jiangsu Canal in China [10], the Ergene River in Turkey [11], the Osun River in Nigeria [12], the Chao Phraya River in Thailand [13], and the Kahaya River in Indonesia [14], etc.

Krueng Aceh River stretches across the Aceh Besar Regency and the Municipality of Banda Aceh. In addition, it is often used as a source for the production of clean water by two regional drinking water companies in Aceh Besar and Banda Aceh. The Krueng Aceh River is also used as a fishing ground by local residents. Despite these benefits, the community has not properly maintained and managed the river, as evidenced by the discovery of plastic waste. This condition raises concerns among environmental observers, particularly particularly for water environments. Therefore, this study identified early pollution in the water of the Krueng Aceh River, particularly by focusing on the effects of MP pollutants.

MP pollution in clean water is a significant environmental health issue that requires special attention from the government, research institutions, and the public. The removal of these materials from raw water to produce clean water has been performed using several techniques, including coagulation, flocculation [15], flotation [16], and filtration [17]. Several reports have shown that MPs can be filtered through a filter but not completely removed, leading to the development of rapid sand-filtration techniques combined with adsorption processes [18]. Membrane technology has emerged as a promising alternative for the removal of MPs from water samples. The use of this method was reported by Pizzichetti et al. using commercial membranes such as polycarbonate (PC), cellulose acetate (CA), and polytetrafluoroethylene (PTFE). MPs in water samples can be effectively removed, but particles in the range of 20-300 μm pass through membrane pores and accumulate inside the permeate tank [19]. To overcome this challenge, high MP removal can be achieved using ceramic membranes [20]. However, filtration using a ceramic membrane is energy-intensive and requires high operating pressures above the average pressure of filtration processes with polymer membrane [21].

In this study, a polymeric membrane was modified to enhance the MP removal efficiency during the cross-flow filtration process. The innovation of modifying membrane structures is a highly important and urgent as an exploration for alternative superior technology to reduce MP pollution in raw water. Membrane filtration with modified pore structures using graphene oxide particles. The obtained membrane was then applied for the removal of MPs in surface water, which served as a source for producing clean water in Banda Aceh/Aceh Besar City, namely, the Krueng Aceh River. Therefore, this study aimed to evaluate the potential for MP contamination in surface water (river water) in Banda Aceh, Saudi Arabia, and to investigate techniques for separating MP particles using PES/GO-modified membrane technology.

2. METHOD

2.1 Material

Polyether sulfone (PES), dimethyl formamide (DMF), and graphene oxide (GO) were obtained from Sigma-Aldrich. Deionized water was supplied by the Environmental Quality Testing Laboratory, Chemical Engineering Department, USK.

2.2 River water sampling

River water sampling was conducted using the integrated sampling method, which combines several samples from a specific location, within the same volume (1000 mL). Sampling points were selected from five different site, representing various segments (Figure 1).

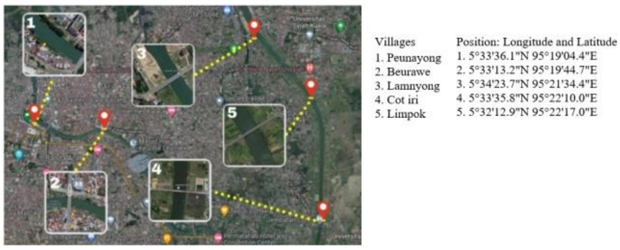


Figure 1. Sampling points in the Krueng Aceh River, Banda Aceh City

Points 1 and 2 represented the urban segment, point 3 represented the estuary, while points 4 and 5 represented residential areas. Points 1 and 2 were chosen to observe whether there is microplastic contamination in urban areas due to garbage disposed into the river or its surroundings, as well as residential areas at sample points 4 and 5. Sample point 3 was chosen because the estuary area is often used for fishing and netting, so it is possible that discarded fishing lines or nets accidentally thrown into the river, can result microplastics over time. At each location, water samples were collected at the same depth, about 1 meter from the surface of the river and stored in labeled sampling bottles.

Before filtering using the membrane, the samples were initially treated with hydrogen peroxide (H_2O_2) and ferrous sulfate heptahydrate ($FeSO_4 \cdot 7H_2O$) to remove organic compounds in the water. 20 mL each of H_2O_2 and $FeSO_4 \cdot 7H_2O$ were added to a water sample, heated for 30 minutes, and then allowed to stand for 1 hour to let sediment settle. The water and sediment were separated from the water. The water was then filtered through 0.45-mm filter paper, and the residue was used for investigating MPs [22, 23].

2.3 Identification of MP concentrations and forms

A Zeiss Primo Star binocular microscope was used to visually analyze, observe, and identify the number and shape of MPs in river water samples before and after the membrane filtration process. Samples were taken using tweezers and placed on a slide, then magnified to 400-1000x until the microplastic particles were clearly visible. The analysis was conducted at room temperature, and all equipment used was sterile. The number and shapes of the MP particles in the photographs were counted, and their dimensions were measured and observed visually. For MP abundance, the parameter was the number of particles per liter. The MP concentration was calculated using Eq. (1) [24].

$$\text{Concentration MPs} = \frac{\text{Total MP Particles counted (particles)}}{\text{Sample Volume (L)}} \quad (1)$$

2.4 Identification of MP types

A Fourier Transform Infrared (FTIR) spectroscopy was performed to identify the type of MPs in water sample before and after filtrated by ultrafiltration module. FTIR is able to detect microplastics by analyzing the unique vibrational spectra of each polymer, enabling the identification of various plastics such as polypropylene and polyethylene in environmental samples. In addition, FTIR provides information on the chemical compound of microplastics, which is important for understanding their degradation processes and potential risks [25]. The spectrum was recorded in the wavenumber range of 400-4000 cm^{-1} with a resolution of 2 cm^{-1} [24].

2.5 Membrane filtration

A cross-flow ultrafiltration module was designed to separate MPs from water samples. A schematic of the ultrafiltration procedure is presented in Figure 2. Two types of flat sheet membranes were placed in the cross-flow module (Figure 2). The advantages of using a cross-flow module include the continuous feed flow across the membrane surface, which prevents the accumulation of foulants on the membrane, the ability to operate continuously, and easier operation compared to dead-end modules. Membranes used in this study are Polyethersulfone (PES) 17 wt% and Polyethersulfone modified with the addition of graphene oxide 0.5 wt% (PES-GO), and the specifications of the membranes are summarized in Table 1.

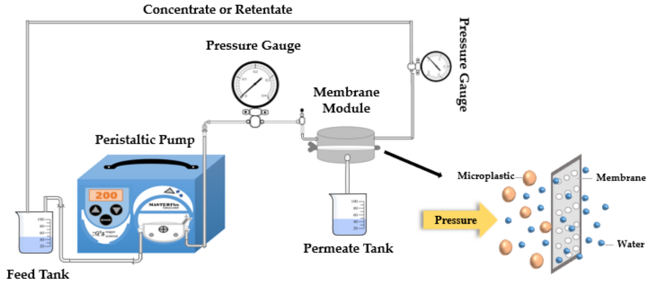
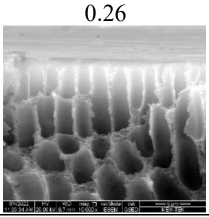
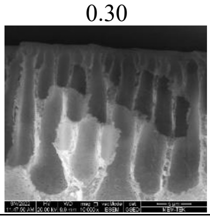


Figure 2. Schematic ultrafiltration equipment for MP removal

The ultrafiltration module (MasterFlex L/S 7518-10, Germany) operates by passing river water, suspected of containing MPs, through membrane layers using a peristaltic pump in a cross-flow filtration setup, as shown in Figure 2. Ultrafiltration for each membrane was conducted at room temperature for 30 minutes at an operating pressure of 0.5 atm. The water that passed through the membrane (permeate) during the 30 minutes filtration process was collected in 10 mL samples for MP content analysis. The MP content in the water before and after filtration was measured using a light binocular microscope.

Table 1. Membrane characteristics

Characteristics	Membrane Type	
	P	**P-GO**
Materials	Polyethersulfon	Polyethersulfon and Graphene Oxide
Water contact angle, Dec	80	68
Tensile strength (MPa)	0.65	0.79
Porosity (%)	58	65
Pore size (μm)	0.26	0.30
Membrane structure		

2.6 Microplastic removal

The efficiency of MP particle removal from water via the membrane separation process was calculated by comparing the number of MP particles in the feed water to the number of MP particles in the permeate, as described by Eq. (2).

$$\eta\ (\%) = (1 - (\frac{M_f}{M_p}))\ x\ 100\ \% \quad (2)$$

where, η was the MP rejection efficiency (%), Mf is the concentration of MPs in the feed (MPs/L), and Mp is the concentration of MPs in the permeate (MPs/L) [26].

3. RESULT AND DISCUSSION

3.1 Total MP in water samples

The total number of MP particles found varied from one sampling point to another. Generally, the sizes of the MP particles found in the water samples of the Krueng Aceh River, Banda Aceh, and Aceh Besar ranged from 0.17 to 60.55 μm. These sizes were confirmed for the MP category. The total number and size of the MPs detected at all sampling locations are detailed in Table 2.

In general, all river water samples were contaminated with MPs, with particle counts per mL of water ranging from 17 to 22. From Table 2, it can be observed that the highest total amount of MP particles was found in the Cot Iri and Lamnyong river water, with a total of 22 particles/mL, followed by those in the Peunayong and Limpok rivers, with 19 particles/mL each. The abundance of MPs found in river water samples may be influenced by the activities of residents around the river.

Table 2. Total and dimensional sizes of MP particles in river water samples from nine sampling locations

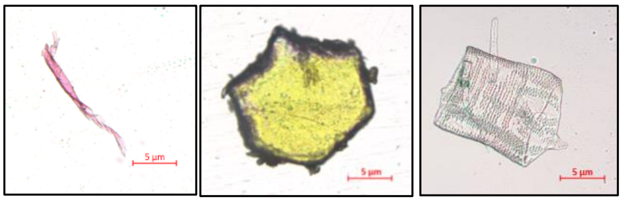
Sampling Location Point	MP Particles	Long (mm)	Wide (mm)
Peunayong	1	18.98	10.67
	2	11.93	5.62
	3	27.38	1.32
	4	35.18	18.69
	5	7.72	6.50
	6	60.40	0.88
	7	10.45	6.67
	8	15.30	13.50
	9	9.30	6.32
	10	14.03	3.01
	11	18.43	18.61
	12	21.15	5.09
	13	15.20	1.25
	14	33.32	1.30
	15	58.93	0.18
	16	20.89	1.62
	17	41.56	0.44
	18	16.55	6.95
	19	13.87	13.30
	20	8.62	8.16
Beurawe	1	12.25	0.61
	2	14.61	1.22
	3	6.40	5.15
	4	20.49	1.11
	5	28.72	2.12
	6	5.60	3.02
	7	44.23	1.06
	8	21.81	1.58
	9	3.80	2.23
	10	6.91	2.59
	11	6.84	7.87
	12	12.23	4.58
	13	11.26	6.19
	14	8.65	6.79
	15	18.98	1.02
	16	8.46	5.85
	17	9.90	5.88
	18	13.74	1.23
	19	12.40	2.16
Cot Iri	1	24.39	0.91
	2	18.66	3.66
	3	52.61	1.58
	4	13.12	6.58
	5	28.72	0.88
	6	6.96	1.63
	7	9.00	11.13
	8	53.58	1.13
	9	11.36	1.29
	10	4.96	2.73
	11	19.57	2.31
	12	9.43	3.05
	13	41.43	0.85
	14	6.61	5.68
	15	35.50	1.28
	16	20.97	0.63
	17	8.41	7.13
	18	9.84	1.12
	19	11.13	9.44
	20	12.83	4.81
	21	11.69	0.85
	22	24.64	1.43
Limpok	1	24.02	1.09
	2	9.88	0.74
	3	24.50	1.41
	4	17.11	0.94
	5	14.29	5.45
	6	18.97	3.00
	7	29.61	2.17
	8	8.05	1.95
	9	20.53	0.94
	10	16.35	1.02
	11	17.52	0.72
	12	9.48	1.62
	13	23.24	1.52
	14	55.69	1.50
	15	36.84	0.52
	16	19.06	5.48
	17	52.43	1.22
	18	13.03	6.75
	19	28.91	1.24
Lamnyong	1	6.23	4.76
	2	21.34	1.37
	3	18.92	1.22
	4	12.16	0.96
	5	18.25	7.11
	6	41.50	1.24
	7	22.86	0.97
	8	16.24	0.44
	9	12.50	10.89
	10	9.56	0.90
	11	16.43	0.79
	12	10.45	1.33
	13	11.24	1.26
	14	18.50	1.37
	15	5.79	5.67
	16	10.90	8.49
	17	15.28	0.98
	18	15.02	1.76
	19	14.38	4.41
	20	15.88	1.17
	21	14.38	6.01
	22	41.30	5.46

3.2 Shape of MP particles

Table 3. MP particles found in Krueng Aceh River water samples

No.	Sampling Point	Shape
1	Peunayong	Fiber, Fragmen, Colorful Fragmen, and Film
2	Beurawe	Fiber, Colorful Fiber, Fragments, Colorful Fragments, and Film
3	Cot Iri	Fiber, Fragment, and Film
4	Limpok	Fiber, Colorful Fiber, Fragments, and Film
5	Lamnyong	Fiber, Colorful Fiber, Fragments, and Film

Three general forms of MP particles were identified: fibers, fragments, and films. As shown in Table 3, the most commonly found type was fiber with a long and thin shape, resembling synthetic fibers. These particles are likely derived from fishing materials such as nets and fishing lines. Fishing activities in the Krueng Aceh River were the main factors that contributed to the abundance of fiber-type MPs [27]. Fragments were larger plastic pieces, while films had thinner layers than fragments. Fragments and films can originate from waste generated by residents around the river, such as plastic bottles, bags, and single-use plastic cups [28]. The three forms of MP particles commonly found in all Krueng Aceh River water samples are presented in Figure 3.



(a) (b) (c)

Figure 3. MP form of the sample before filtration: (a) colored fiber, (b) colored fragment, and (c) Film

3.3 Microplastic particle types

Qualitative analysis was conducted to confirm the types of MPs present in the river water samples by identifying the functional groups of compounds or substances in the river water samples using FTIR [29]. The FTIR spectra of the river water samples were analyzed using several standard polymer spectra. The functional groups in the IR spectra of the Krueng Aceh River water samples are illustrated in Figure 4.

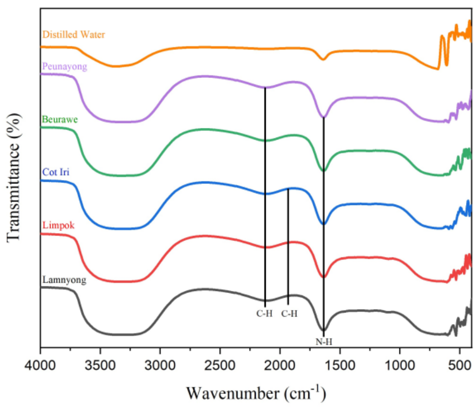


Figure 4. Functional group analysis of river water samples

Table 4. MP particle types based on IR spectra analysis

No.	Sample	Wavenumber Wave (cm^{-1})	Bond	MP Particle Types
1	Peunayong	2368.59	C-H	PE
		2117.84	C-H	PE
		1901.81	C-H	PE
		1635.64	C=O	PA
2	Beurawe	2115.91	C-H	PE
		1635.64	C=O	PA
3	Cot Iri	2115.91	C-H	PE
		1635.64	C=O	PA
4	Limpok	2115.91	C-H	PE
		1863.24	C-H	PP
		1635.64	C=O	PA
5	Lamnyong	2113.98	C-H	PE
		1863.24	C-H	PP
		1635.64	C=O	PA

*PE (*Polyethylene*), PA (*Polyamida*), PP (*Polypropylene*)

The interpretation of the IR spectra depicted in Figure 4 used standard polymer spectra, allowing identification of the functional groups present in the river water samples, as outlined in Table 4. The FTIR analysis of the river water samples revealed bonds close to the standard wavenumber of polyethylene, polypropylene, and polyamide polymers. The presence of polyethylene was evidenced by the IR peaks observed at wavenumber 1901.81; 2113.98; 2115.91; 2117.84; and 2368.69 cm^{-1}. This polymer is commonly used for manufacturing plastic bags. The presence of polypropylene was indicated by an IR peak at wavenumber 1635.64 cm^{-1}. Polypropylene MPs originated from straw and bottle caps. Furthermore, the presence of the polyamide polymer was indicated by an IR peak at wavenumber 1635.64 cm^{-1}. Polyamide is a major component of nylon and is typically used in the production of fabric fibers and fishing nets [30].

In general, the types of microplastics found at the five sample points did not show significant differences. The Peunayong sample was dominated by PE, indicating microplastics from plastic bags, presumably due to the large number of single-use plastic bags discarded after purchasing food or goods in urban areas, similar to the Beurawe sample point. At the Limpok sample point, PP microplastics were found, indicating microplastics from straws and drink bottles, likely due to the disposal of straws from children's snacks in residential areas. The PA microplastics found in the river are thought to originate from fishing activities using hooks or nets, but they could also be caused by residents' washing activities. Based on these results, it can be concluded that the types of microplastics found are strongly influenced by the surrounding environment and activities.

3.4 Abundance of MPs after filtering

3.4.1 Total particle size and size after ultrafiltration

The total number and size of MP particles remaining in the water samples after ultrafiltration by both types of membranes is summarized in Table 5. Generally, the number and size of the MP particles decreased significantly. The average number of MP particles in the river water samples before ultrafiltration was 18.6 particles/mL, whereas the average number of MP particles after ultrafiltration with the P membrane and P-GO membrane decreased to 4.6 and 5.4 particles/mL. The filtration process using membrane technology relies heavily on the pore structure of the membrane [31-33]. The porosities and pore sizes of the P-GO membranes were slightly higher than the P-GO membrane. Therefore, the number of MPs remaining in the water sample after filtration with the P-GO membrane was slightly larger than that with the P membrane.

3.4.2 Formation of MP particles in the permeate

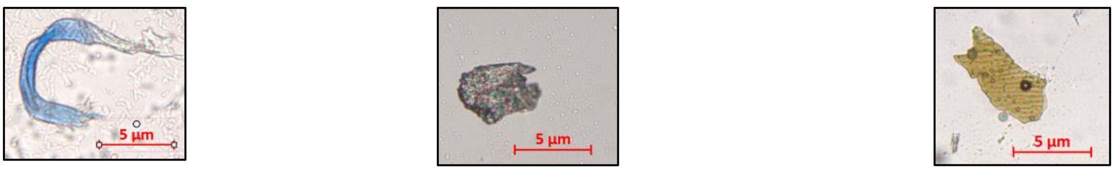
The MP particles present in the permeate after 30 min of ultrafiltration at an operating pressure of 1 atm are summarized in Table 6. Generally, the MP particles found in the permeate had similar shapes to those in the ultrafiltration feed sample, but they varied in length and width, as illustrated in Figure 5. The MP particles identified in the permeate were smaller in number and size than those found in the ultrafiltration feed sample, as shown in Figure 3. MP particles with diameters larger than the membrane pores could not pass through the ultrafiltration module. Therefore, some forms of these particles that were initially found in the river water samples were no longer present in the permeate [34]. For example, in the river water from the sampling location of Beurre, MPs of fibers, colorful fibers, fragments, colorful fragments, and films were detected. After ultrafiltration through the P membrane, only the MPs fiber and film remained in the permeate. Similarly, in the water sample taken from Anong, only MPs fibers and films were found after ultrafiltration through the P membrane.

Table 5. Total and size of MPs in river water after ultrafiltration

Membrane	Sampling Location Point	Number of MPs Particles	Long (mm)	Wide (mm)
P	Peunayong	1	13.62	0.99
		2	12.05	0.98
	Beurawe	1	6.88	5.06
		2	9.82	3.70
		3	7.06	0.97
	Cot Iri	1	8.35	7.31
		2	8.06	5.44
	Limpok	1	8.77	0.87
		2	9.15	1.58
	Lamnyong	1	8.37	2.93
		2	8.51	7.84
		3	4.20	2.99
P-GO	Peunayong	1	10.38	0.80
		2	7.90	1.35
		3	5.18	3.34
	Beurawe	1	8.79	1.49
		2	6.46	5.90
		3	7.47	5.09
	Cot Iri	1	5.65	0.81
		2	12.98	1.40
		3	9.17	1.27
	Limpok	1	9.69	4.58
		2	4.47	4.10
	Lamnyong	1	9.63	1.10
		2	9.93	6.12
		3	9.04	1.83

Table 6. Formation of Mp particles in the permeate

Sampling Point	Membrane	Shape
Peunayong	P	Fiber, Colorful Fragments, and Film
	P-GO	Fiber, Colorful Fragments, and Fragments
Beurawe	P	Fiber and Film
	P-GO	Fiber, Colorful Fiber, Fragments, and Film
Cot Iri	P	Fiber, Colorful Fragments, and Film
	P-GO	Fiber and Colorful Fibers
Limpok	P	Fiber, Colorful Fiber, and Film
	P-GO	Fibers, Colored Fibers, Fragments, and Films
Lamnyong	P	Fiber and Film
	P-GO	Fragments and Movies



(a) (b) (c)

Figure 5. Forms of MPs contained in permeate (a) Colorful fiber, (b) Film, and (c) Colorful fragments

3.4.3 Rejection of MP particles

The rejection of MPs after filtration by the P- and P-GO membranes was calculated in 2 categories, namely particle number removal and size reduction. The rejection of the particle number was determined by observing the number of particles in the river water samples and permeates. The rejection efficiency of MPs was then calculated using Eq. (2). The particle size rejection was determined by comparing the length of particles found in the river water sample to the length of particles remaining in the permeate. The profiles of MPs particle number and size rejection by the P- and P-GO membrane filtration systems are shown in Figures 6 and 7, respectively.

The rejection of MP particles generally exceeded 85% for all river water samples after passing through the filtration process using both types of membranes. The rejection of MP particles at the Penayong and Cot Iri sampling locations reached 90% after filtration through the P membrane. Figure 7 shows that the size of the remaining MP particles in the permeate was approximately 90% smaller than the size of the

MP particles in the river water samples. The size of MP particles in the river water samples originating from the Cot Iri and Limpok sampling locations could be reduced by up to 96% after filtration through both membrane types. The different membrane characteristics used in this study did not significantly affect the rejection of MP particles.

The rejection coefficient of membranes containing additives usually depends on the type of substance separated during the filtration process. The type of feed solution for filtration, whether it was particles, ions, or compounds, could have different rejection tendencies when using the same membrane [35-37].

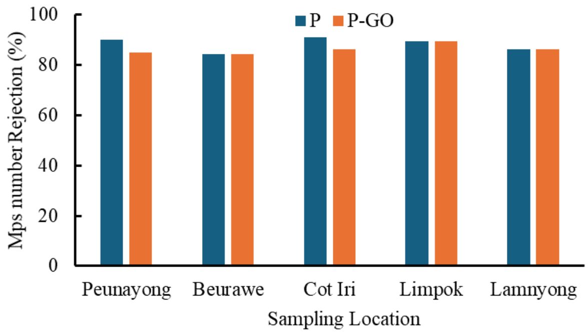


Figure 6. Rejection of the number of MP particles in the river water samples at each sampling location after filtration through the P- and P-GO membranes

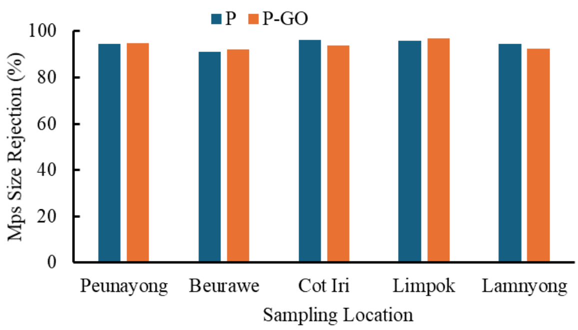


Figure 7. Rejection of MP particle sizes in river water samples from each sampling location after filtration using P- and P-GO membranes

4. CONCLUSIONS

This study investigated the abundance of microplastics in the Krung Aceh River at five different sample points, with an average detection of 17-22 particles/mL, predominantly in the form of fibers, fragments, and films. The results showed that the most commonly found type of microplastic was PE, originating from plastic bags. The separation of microplastics using P and P-GO membranes showed promising results, with a rejection rate of 91% and a particle size reduction of 96.7%. Due to the high separation efficiency achievable with membrane technology, it is expected to be further implemented in urban drinking water filtration systems to address microplastic issues, specifically in Aceh. The high separation efficiency can improve overall water quality, providing positive impacts on ecosystems and the environment.

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NOMENCLATURE

GO	Graphene oxide
MPa	Unit of pressure (Megapascal)
MPs	Microplastics
P	Pristine Polyethersulfone
PES	Polyethersulfone
P-GO	Polyethersulfone modified with graphene oxide