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A laboratory-scale graywater treatment system based on a membrane filtration and oxidation process — characteristics of graywater from a residential complex

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Abstract

If wastewater can be reused, both the demand for water and the amount of wastewater can be reduced. Wastewater originating from all sources in residential homes other than the toilet is defined as graywater. In this study, graywater was treated through a microfiltration membrane and oxidation process, and the efficiency of the removal of color, turbidity, COD, suspended solids, *E. coli*, total coliform, *Salmonella* and *Staphylococcus* was studied. The treated graywater was reused for purposes other than drinking and bathing, such as fire fighting, water for plants, water for toilets and car washing. The pH of the treated graywater was 7–7.7. The removal efficiency of each factor with the microfiltration membrane was as follows: color (98%); turbidity (99%); COD (99%); suspended solids (99%); *E. coli*, total coliform, *Salmonella* and *Staphylococcus* (30%). Following the membrane filtration process, the efficiency of the removal of each factor using the oxidation process was as follows: color (100%); turbidity (99%); COD (99%); suspended solids (99%); *E. coli*, *total coliform*, *Salmonella* and *Staphylococcus* (100%). The quantity and quality of the graywater was sufficient to establish a sustainable water circulation system for the reuse of residential graywater. The A_2O–MF membrane–OP system showed good treatment capacity. The A_2O–MF membrane system could effectively remove COD, turbidity, color, and suspended solids, while OP was effective for the removal of *E. coli, total coliform, Salmonella* and *Staphylococcus*.

Keywords: Graywater; A_2O–MF membrane; Oxidation process; Residential complex

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1. Introduction

Most countries lack sufficient regulations or specific guidelines on the reuse of graywater, as frequently the health risks are overestimated while the environmental risks are underestimated. With the development of industry, urbanization and the diversification of lifestyles, there is currently a high demand for water. In addition to this, with the recent construction of many large-scale multi-unit complexes, there have been problems relating to the increased demand for water and the increased amount of wastewater. Indoor domestic water demand (excluding garden irrigation and other external uses) in developed countries usually ranges between 36 and 66 m^3/y per capita [1–4], comprising 30–70% of the total urban water demand. Aside from a minor quantity, most consumed water is transformed into wastewater, which can be classified into two major categories:

- Graywater: originating from all household “water-generating” appliances except for toilets, comprising 60–70% of the in-house water demand.
- Blackwater: originating from toilets, comprising 30–40% of the in-house water demand.

In urban areas, the most feasible graywater reuse option is water for toilet flushing, which can reduce individual in-house net water demand by 40–60%. If this practice becomes widespread, a reduction of up to 10–25% in urban water demand can be achieved. However, graywater can be quite polluted and thus may pose health risks, as well as having negative aesthetic factors (such as an offensive odor and color) and environmental effects [1,5–7]. Consequently, the reuse of graywater requires highly efficient and reliable conveyance, storage and treatment systems. A wide variety of treatment systems has been reported in the literature, and these vary greatly in complexity and degree of treatment [1,6,8–13]. Ramon et al. [14] studied graywater treatment using an ultrafiltration and nanofiltration membranes. Garland et al. [15] studied graywater treatment using hydroponic plants. Gross et al. [16] studied graywater treatment using a vertical flow constructed wetland. However, all studies concluded that additional disinfection is required for the reuse of graywater. Even when reused for purposes other than drinking and bathing, such as fire fighting, water for plants, water for toilets and car washing, a certain standard of reused water quality must be satisfied. In Korea, the quality standard for reused water is as follows: COD of less than 20 mg/L, turbidity of less than 2 NTU, color less than 20 Pt-Co, suspended solids less than 5 mg/L, and zero *E. coli*.

The objectives of this study are (1) to analyse quantity and quality of graywater, and (2) to evaluate the A_2O (anaerobic– anoxic– oxic)–MF membrane–OP (oxidation process) system to remove COD, turbidity, color, suspended solids, *E. coli*, total coliform, *Salmonella* and *Staphylococcus* for graywater treatment.

2. Materials and method

2.1. Sampling

The area of each apartment in the complex studied was 55.45 m^2, and 170 apartments were studied in total. The apartment complex studied is located at Bundang-Gu, Kyungki-Do, Korea. The quantity and quality of graywater were analyzed once every hour for periods of 24 h at a time. To reflect the characteristics of different days, sampling was performed both on weekdays and on weekends. To reflect seasonal variation, sampling was performed in winter and spring. To evaluate the A_2O–MF membrane–OP system, samplings were taken every day for 30 days.

2.2. Analytical methods

pH was determined using a glass electrode pH meter (Orion, Model 525A). CODcr, turbidity and color were measured using Hach digestion vials (Hach, DR/2012). Measurement of MLSS followed the 2540D method in Standard Methods [17]. *E. coli* and coliform were estimated using the CHROMagar TM ECC. *Salmonella* was estimated using desoxycholate citrate agar. *Staphylococcus* was estimated using Mannitol salt agar. The microbial community was analyzed using DNA extraction and PCR. According to the results of PCR, T-RFLP was used. To detect *E. coli*, coliform, *Salmonella* and *Staphylococcus*, the following method was used: First, the specimen was inoculated directly on the surface of the medium. Second, the plate was incubated at 35± 2°C for 18–24 h. To analyze the microbial community, the Powersoil DNA Isolation Kit (MO BIO) was used for DNA extraction, and MJ mini (BioRad) was used for the detection of PCR. According to results of PCR detection, T-RFLP was performed. Amplification was performed with a pair of universal bacterial primers (27F[5′AGAGTTTGATCATGGCTCAG3′] and 1492R[TACGGTTACCTTGTTACGACTT3′]) for T-RFLP (27F-FAM 5′-FAM-AGA]).

2.3. A_2O–MF membrane–OP system

2.3.1. A_2O–MF membrane system

Fig. 1 represents the experimental system used in this study, which consists of an anaerobic bioreactor, an anoxic bioreactor and an aerobic submerged MF membrane. The volume of the anaerobic bioreactor is 2.5 L. The anoxic bioreactor and oxic bioreactor are connected by circular columns with working volumes of 2 L and 8 L, respectively. The mixed liquid of the oxic bioreactor was returned to the anoxic bioreactor with an internal recycle ratio of 1.5*Q* by a pump. During the 60 days of the experiment, total HRT was maintained at 6 h (1.2 h/anaerobic reactor, 0.96 h/anoxic reactor, 3.84 h/oxic reactor). The *F/M* ratio was maintained at 0.11–0.17 kg BOD/MLVSS/d. MLSS was maintained at 4500 mg/L. The module type of membrane was hollow fiber, and its pore size was 0.4 μm. The material was polyvinyl difluoride. The operation type of membrane was the submerged type, and urethane and epoxy resin were used for bonding. The model of the membrane was SuperMAK®.

2.3.2. Disinfection system

Fig. 2 shows the ozone experimental system used in this study. This process was performed after the graywater had passed through the A_2O–MF membrane system. The ozone reactor consisted of an ozone generator, an ozone detector, an acrylic reactor, equipment for the removal of bubbles, KI solution, and equipment to destroy the remaining ozone. Ozone contacted the graywater in the cylindrical type batch reactor by

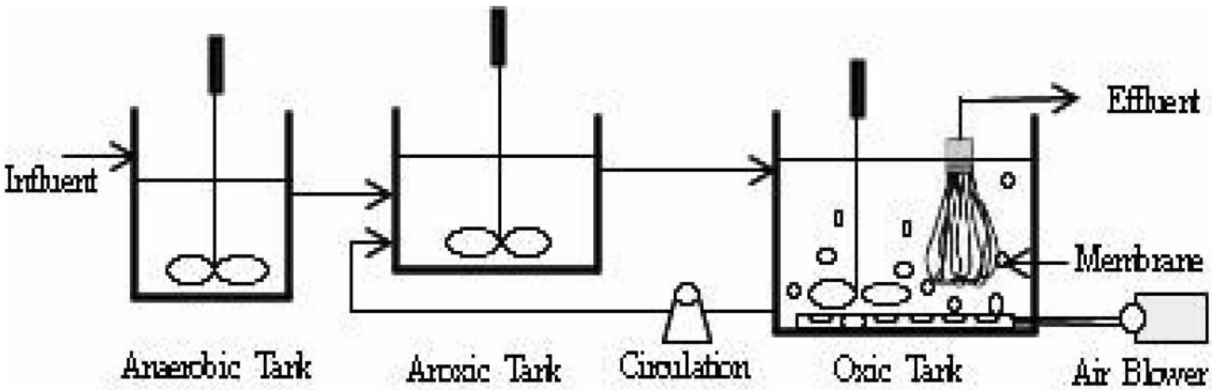


Fig. 1. Schematic diagram of A_2O–MF membrane system.

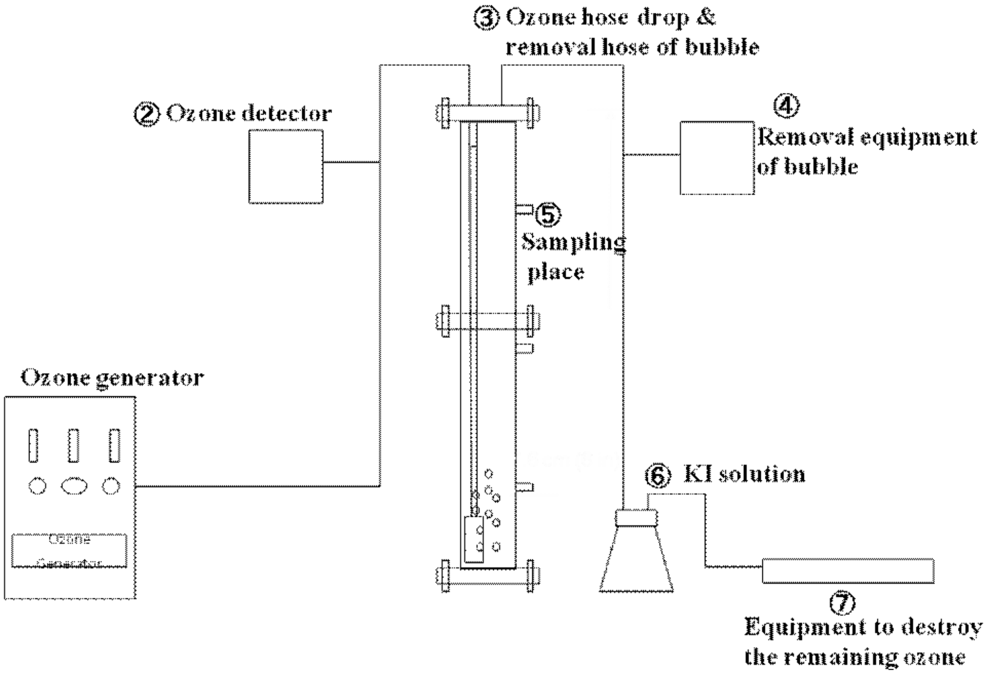


Fig. 2. Schematic diagram of ozone experimental system.

Table 1
Characteristics of ozone concentration according to the O_2 flow rate and output voltage

O_2 flow rate (LPM)	Output voltage (kV)	O_3 amount (g/h)
1.5	6	2.6
	7	4.0
2	6	2.6
	7	4.8
2.5	6	2.4
	7	3.7
	8	5.4
3	7	3.2
	8	5.2

bubbling. The reactor was made of acrylic to avoid oxidation and to allow the easy observation of changes in the water's color. A ceramic air distributor was located at the bottom of the cylinder. Ozone was introduced into the reactor through this distributor.

Ozone was generated by flowing the oxygen into the ozone generator. The ozone concentrations at different O_2 flow rates and output voltages are listed in Table 1. The results of the experiments were estimated at 10 min, 20 min, and 30 min after becoming in contact with ozone. To determine optimal O_2 flow rates and output voltages in terms of the efficiency of the removal of color, turbidity and organic matter, a total of nine experiments were performed, and 2.5 LPM and 7 kV were determined to be the optimal O_2 flow rate and output voltage, respectively.

3. Results and discussion

3.1. Quantity and quality of graywater

Fig. 3(a) and (b) shows the concentration of COD in the raw graywater. The maximum concentration of COD was 400 mg/L, and the average concentration of COD was 50 mg/L, regardless of the season. In the studies previously conducted [14,18–20], the average concentrations of COD were 170 mg/L, 420 mg/L, 70 mg/L and 501 mg/L respectively. It can be seen that there are considerable differences in these concentrations, which may stem from the fact that the measurements were taken in different countries. With regard to the source of the graywater, COD concentration was higher in outflow from the kitchen than from the bathroom. In terms of the time of day, COD concentrations were higher in the afternoon than in the morning. With regard to the differences in the graywater according to the day, COD concentration was high in outflow from kitchen on Sunday in the winter, and in outflow from the kitchen on weekdays in spring.

Fig. 3(c) and (d) shows the turbidity of the raw graywater. In winter, the maximum turbidity was 150 NTU, and the average turbidity was 60 NTU. In spring, the maximum turbidity was 200 NTU, and the average turbidity was 10 NTU. In studies previously conducted [14,18,20], the average turbidities were 23 NTU, 84.8 NTU and 19 NTU, respectively. With regard to source and day, the turbidity of the outflow was high on weekdays in winter, while the turbidity of the outflow from the bathroom was high on Saturdays in spring. In terms of time, turbidity values were high between 18:00 and 20:00 in winter.

Fig. 3(e) and (f) shows the color of the raw graywater. In winter, the maximum value of the color was 130 Pt-Co, and the average value of color was 40 Pt-Co. In spring, the maximum concentration of color was 115 Pt-Co, while the average concentration of color was 10 Pt-Co. With regard to source and day, the concentration of color was high in the outflow on weekends in winter, and in the outflow from the kitchen on Saturdays in winter. In terms of time, the concentration of color was high from 18:00–20:00 in the winter.

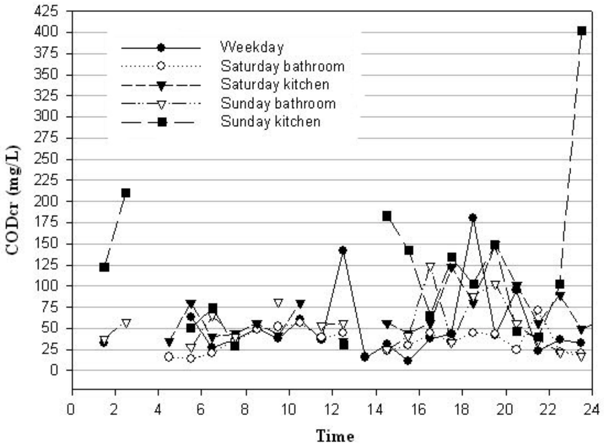
Fig. 3(g) and (h) shows the concentrations of suspended solids in the raw graywater. In winter, the maximum value of suspended solids was 120 mg/L, and the average value of suspended solids was 40 mg/L. In spring, the maximum value of suspended solids was 130 mg/L, and the average value of suspended solids was 30 mg/L. With regard to source and day, outflow concentration of suspended solids on weekdays was high in winter, while outflow concentration of suspended solids from the bathroom was high in winter on Saturdays. In terms of time, concentrations were high from 18:00–22:00.

Fig. 3(i) and (j) shows the quantity of outflow by season, day, source, and time. Maximum quantity of outflow was 0.3 m^3/h in winter. In spring, the maximum quantity of outflow was 1.3 m^3/h. With regard to time, the quantity of outflow was high from 6:00–8:00 and 18:00–22:00 regardless of the season.

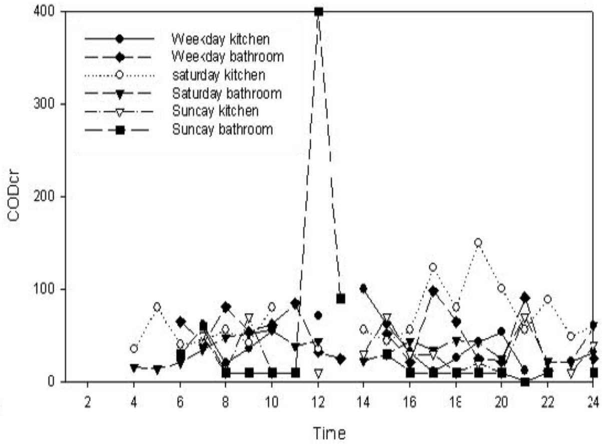
3.2. Evaluation of A_2O–MF membrane system

Fig. 4(a) shows the efficiency of COD removal by the A_2O–MF membrane system. Its maximum value in the influent was 1600 mg/L (29th day), and the minimum value of influent was 42 mg/L. The difference in the concentration of each factor in influent between Fig. 3 and Fig. 4 can be examined, because the data in Fig. 3 show a measurement taken every hour for a day, while the data in Fig. 4 show a measurement taken every day for 30 days. Removal efficiency of COD was about 75%–100%. Although the removal efficiency was 75% at the 5th day, the actual level of COD in effluent was 12 mg/L. As the legal standard for water reuse is less than 20 mg COD/L in Korea, this value satisfies the legal standard. By the 15th day, the efficiency of COD removal was 100%.

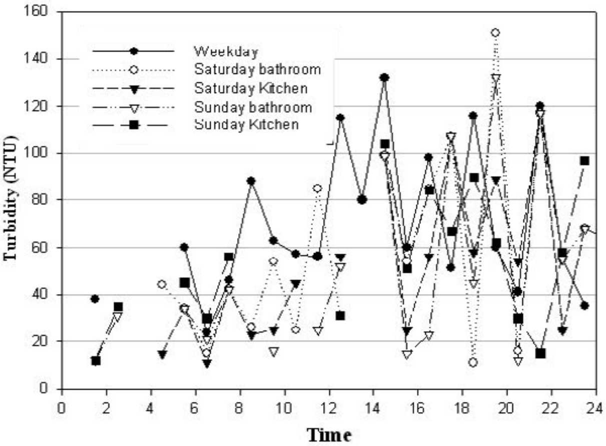
Ramon et al. [14] conducted experiments of low strength graywater treatment using both ultrafiltration and nanofiltration membranes. Depending on the membrane used, UF removed 45–70% of COD as well as 92–97% of turbidity. Depending on the membrane used, NF removed 93.3% of COD as well as 98.1% of turbidity. Fig. 4(b) and (c) shows the efficiency of the



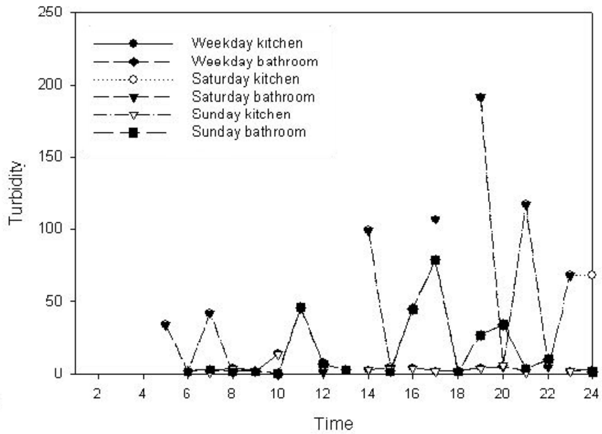
(a) COD in Winter



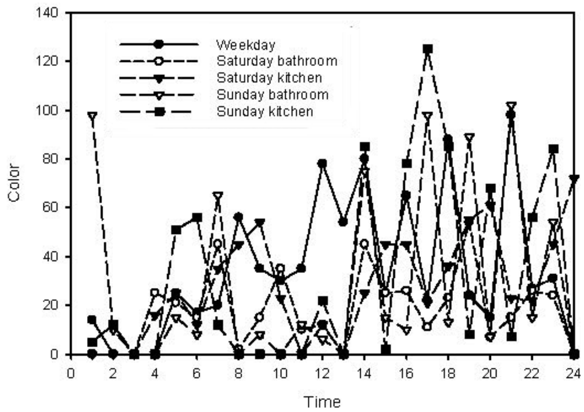
(b) COD in Spring



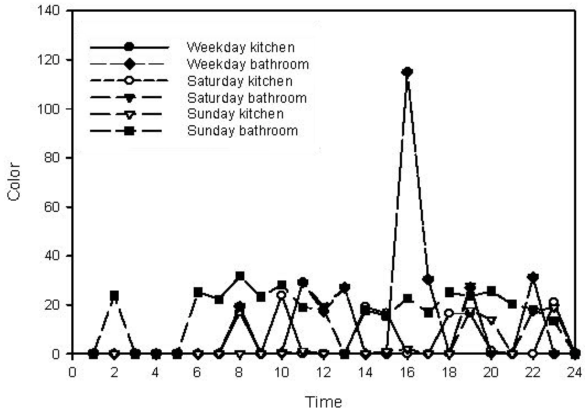
(c) Turbidity in Winter



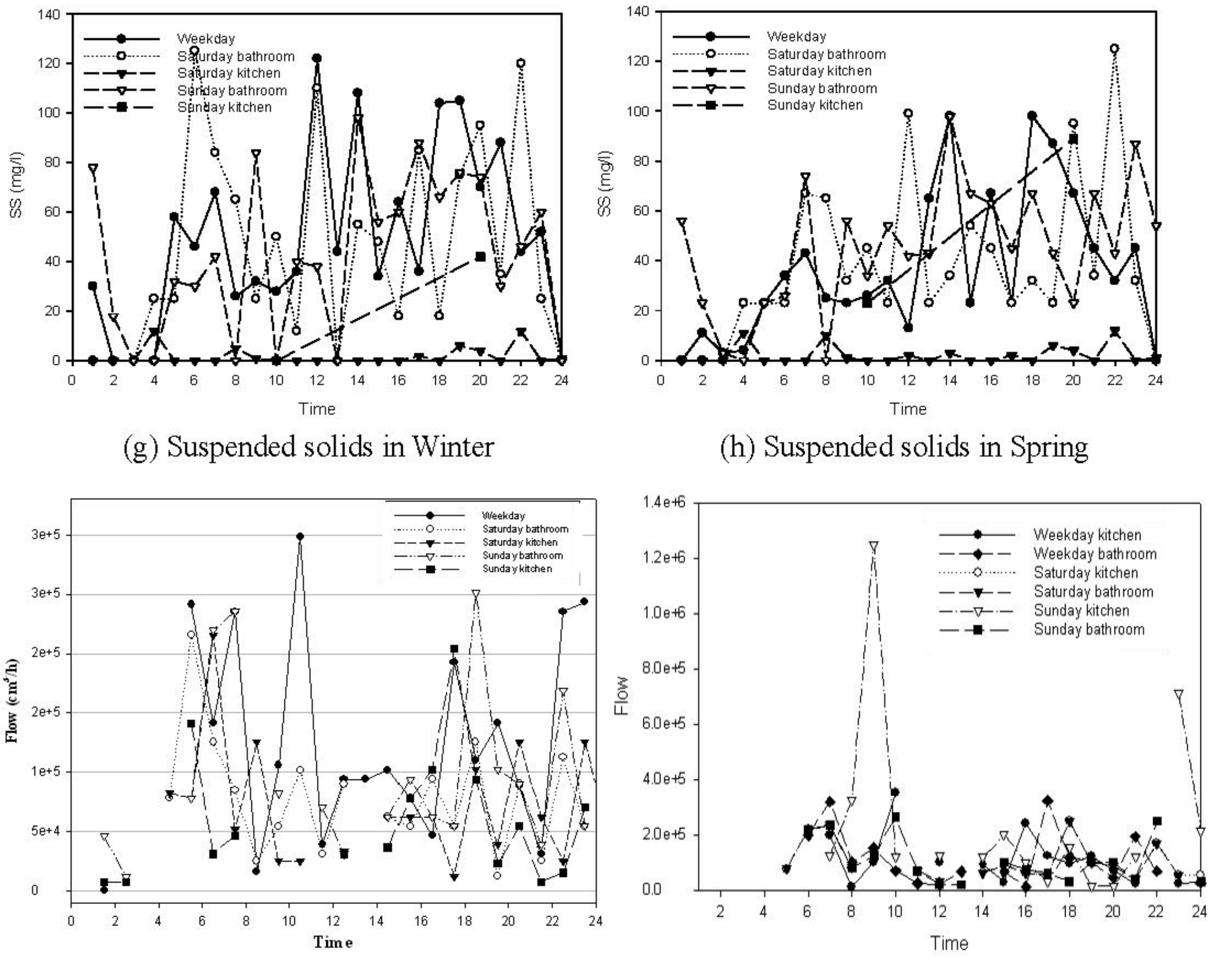
(d) Turbidity in Spring



(e) Color in Winter



(f) Color in Spring



(g) Suspended solids in Winter

(h) Suspended solids in Spring

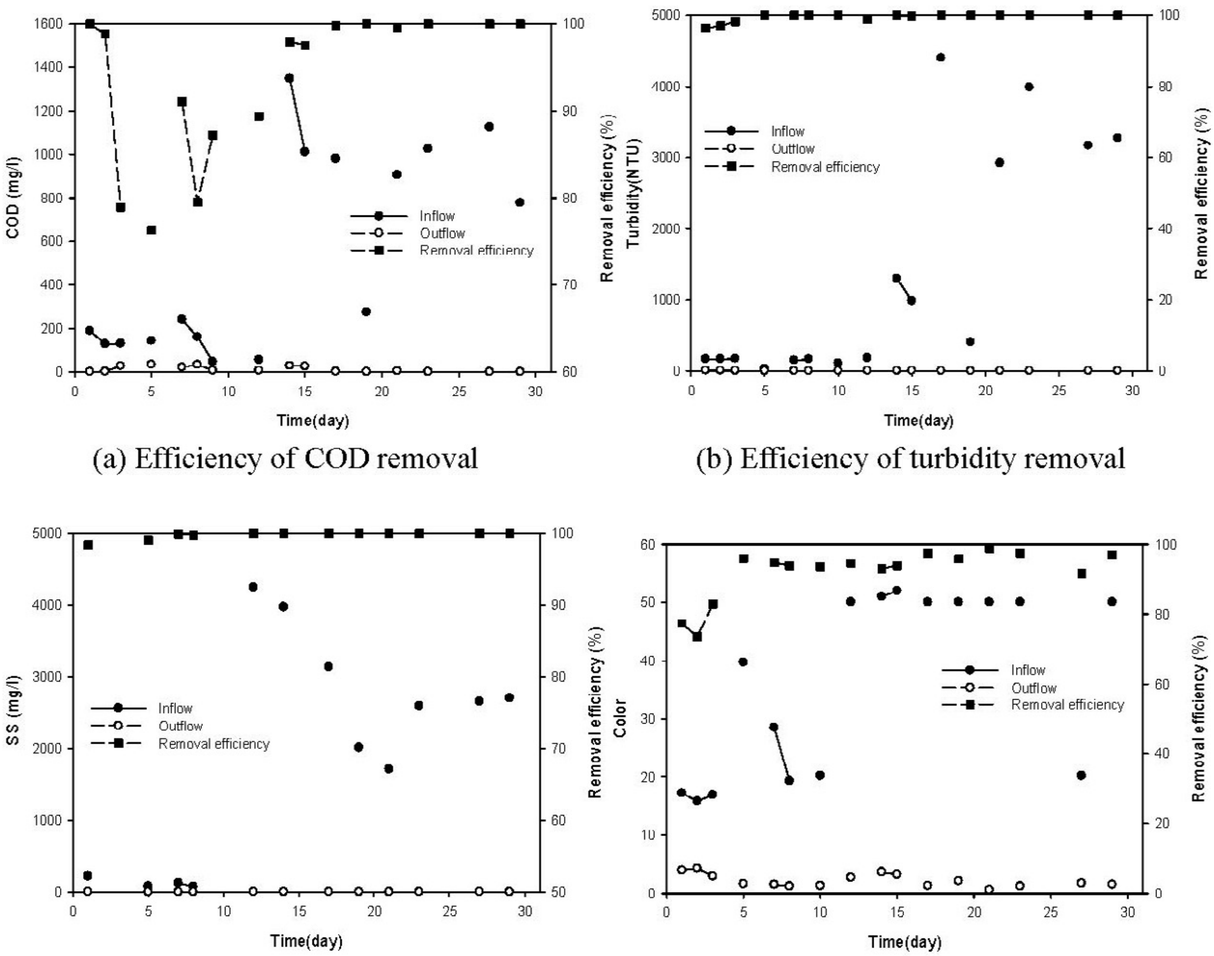
(i) Quantity of outflow in Winter

(j) Quantity of outflow in Spring

Fig. 3. Analysis result of COD, turbidity, color, suspended solids and quantity of outflow. (The lack of a line connecting two points on the graph indicates that consecutive sampling was not performed).

removal of turbidity and suspended solids by the A_2O–MF membrane system. Maximum value of influent was 4,300 NTU, and the minimum value of influent was 31 NTU. Maximum value of suspended solids in the influent was 4,200 mg/L, while the minimum value was 72 mg/L. Due to the activated sludge and MF membrane, suspended solids were removed very effectively. The efficiency of turbidity removal was 98–100%. Fouling of the MF membrane did not occur in this study. Fouling is a major issue in the membrane process. For example, Lodge et al. [3] compared wastewater with graywater in UF following biological treatment and found similar fouling behavior, reasoning that the difference could be explained with a higher SS concentration in wastewater in that case. In this study, because suspended solids were removed effectively using the A_2O process, it seems that hardly any fouling of the MF membrane occurred.

Fig. 4(d) shows the efficiency of color removal by the A_2O–MF membrane system.



(a) Efficiency of COD removal

(b) Efficiency of turbidity removal

(c) Efficiency of suspended solids removal

(d) Efficiency of color removal

Fig. 4. Efficiency of COD, turbidity, suspended solids and color removal. (The lack of a line connecting two points on the graph indicates that consecutive sampling has not been performed).

Maximum value of influent was 51 Pt-Co, and the minimum value of influent was 15.8 Pt-Co. As a result of the activated sludge and MF membrane, organic matter, suspended sludge and dissolved organic matter (DOM) were removed effectively. Efficiency of color removal was greater than 90% from the 5th day.

The decolorizing of wastewater using various membrane-based pressure-driven processes has been investigated by several researchers [21–24]. MF and UF are low pressure membrane processes that separate suspended solids, macromolecules and colloids from a feed stream [4,25]. As a pretreatment step, MF can significantly enhance water quality for a reasonable cost. Decolorizing textile wastewater [8], municipal wastewater and groundwater supplies [26] are some examples of the applications of membranes in this area.

Table 2 shows the detection of micro-organisms in influent, oxic reactor and effluent. The A_2O–MF membrane system could not completely remove *E. coli*, total coliform, *Salmonella*

Table 2
Detection of microorganisms in influent, oxic reactor, and effluent (unit: CFU/100 mL)

	All microorganisms	*E. coli*	Coliform	*Staphylococcus aureus*	*Salmonella tyohimurium*
Influent	583,200	4,000	1,200	1,800	5,400
Oxic reactor	580,000	3,200	900	1,700	5,100
Effluent	563,000	1,300	700	1,300	4,300

and *Staphylococcus* from the graywater, regardless of concentration. Pathogenic microorganisms were detected in effluent. Generally, the size of *E. coli* is 0.5×1.5 µm, while that of *Staphylococcus* is 1 µm. The MF membrane could not completely remove pathogenic microorganisms. Although the number of pathogenic microorganisms was decreased, the MF membrane could not achieve zero detection of *E. coli*, which is the standard for water reuse.

3.3. Evaluation of the oxidation process

Fig. 5 shows the removal tendency of COD, turbidity, color, and SS according to residence time in the ozone reactor. The values on the graph are the means of total experimental values using OP. At 0 min, although graywater was treated by the A_2O–MF membrane system, the concentration of each factor was not good (more than the accepted standard for water reuse in Korea). Because the A_2O–MF membrane system showed insufficient removal efficiency until the 5th day of the experiment, the mean of all experimental values did not meet the standard for water reuse in Korea. At 10 min, the concentration of color decreased from 40 Pt-Co to 20 Pt-Co and that of COD was decreased from 44 mg/L to 14 mg/L, turbidity was decreased from 14 NTU to 3 NTU and suspended solids decreased from 20 mg/L to 5 mg/L. At the range of 0–15 min, reduction of each factor was almost performed. These results have already been reported in the literature [13,15]. At 20 min, the concentration of color was slightly changed, and the other factors showed similar results considering the margin of error. At 30 min, concentration of suspended solids was slightly changed, and the other factors showed almost identical removal performance.

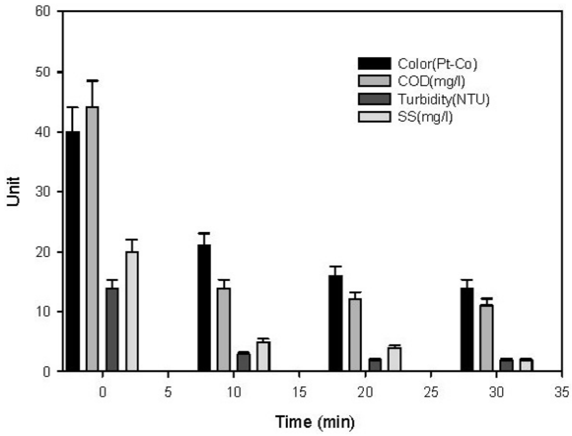


Fig. 5. Effluent concentration as a function of residence time in the ozone reactor.

The efficiency of *E. coli*, total coliform, *Salmonella* and *Staphylococcus* removal was 100%. Although the A_2O–MF membrane system could not efficiently remove them, the OP unit did efficiently remove *E. coli*, total coliform, *Salmonella* and *Staphylococcus*. In addition, all pathogenic microorganisms were removed at the range of 0–15 min.

4. Conclusions

Graywater is often used without any significant pretreatment, a practice that is mistakenly considered safe. The results reported in this study indicate that an A_2O–MF membrane–OP system has good treatment capacity. The A_2O–MF membrane system studied could effectively remove COD, turbidity, color, and suspended solids, while the OP was effective for the removal of *E. coli*, total coliform, *Salmonella* and *Staphylococcus*. As the MF membrane could not completely remove E. coli, total coliform, *Salmonella* and *Staphylococcus*, a disinfection system was required.

If graywater is reused for human contact, a disinfection system has to be installed. The A_2O–MF membrane–OP system satisfies the Korean standards for the reuse of water, which are: COD of less than 20 mg/L, turbidity of less than 2 NTU, color concentration of less than 20 Pt-Co, suspended solids of less than 5 mg/L, and zero detection of *E. coli*. Since the MF membrane was very effective for the treatment of COD, turbidity, color and suspended solids, it was unnecessary to use UF or NF membranes. The MF membrane was also more economic than either the UF or NF membrane.

Friedler et al. [13] reported that if the practice of on-site graywater reuse becomes widespread, the costs of the systems will obviously decrease, making them more appealing to individual consumers. In addition, under typical conditions, on-site graywater reuse is a feasible solution for decreasing overall urban water demand, not only from an environmental standpoint, but also in economic terms. Treated graywater can be used for toilets, fire fighting, car washing, green spaces, and for constructed wetlands and ponds. Prathaper et al. [27] reported that it would be beneficial to individuals and to society if the building industry could be persuaded to install graywater treatment systems in (a) new houses; (b) new apartment complexes; and (c) public buildings, such as mosques and schools, where existing plumbing could easily be modified to separate graywater from blackwater. Ultimately, a sustainable water circulation system can be accomplished.

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