PRE-TREATMENT OPTIONS FOR HYBRID CERAMIC MICROFILTRATION SYSTEMS FOR SURFACE WATER TREATMENT

Amila Abeynayaka[a*], Le Anh Tuan[a], Chettiyappan Visvanathan[a], Hitoshi Yonekawa[b], Yoshimasa Watanabe[c]

[a] Environmental Engineering and Management Program, Asian Institute of Technology, P.O. Box 4, Klong Luang, Pathumthani 12120, Thailand

[b] Membrane Engineering Department, Business Development Division, MetaWater, Nagoya 467-8530, Japan

[c] Division of Built Environment, Graduate School of Engineering, Hokkaido University, Sapporo 060-8628, Japan

*Corresponding author Tel.: +66 2 524 6189; fax: +66 2 524 5625. E-mail address: amila@ait.ac.th

Abstract
Membrane filtration is one of the alternatives suggested for the future drinking water production. Worldwide, the total number of ceramic membrane usage for water treatment is increasing swiftly and this is the first ceramic filtration study which is applied to a surface water treatment in a tropical region. This study was conducted to investigate the removals of natural organic matter and pathogens and, the fouling behaviour of the ceramic microfiltration system (CMF) when combined with the pre-treatments namely coagulation-flocculation and adsorption. Further, this study was carried out for three different scenarios namely: direct CMF (Scenario 1), coagulation-flocculation (PACl) CMF (Scenario 2), and coagulation-flocculation (PACl) and adsorption (PAC) CMF (Scenario 3). The outcome of the study revealed that the suspended solid, total coliform and fecal coliform were removed completely in all three scenarios. However, the removal efficiency on *Giardia* and *Cryptosporidium* was the highest (99.92%) with scenario 3 when compared with that of the other two scenarios. In addition, both the highest TOC and DOC removals were noted as above 80% with scenario 3. The fouling behaviour of the system varied with the scenarios, and long filtration period and high TMP recovery were achieved with the third scenario.

Key words: Surface water; Ceramic membrane; Microfiltration; Coagulation-flocculation; Adsorption.

1. Introduction

The outburst of population and the spread of industrialization whip the integrative adverse impact on the environment in the forms of air pollution, water pollution, ozone depletion, climate change, food contamination, poor sanitation, etc. The wayward activities of human kind have seriously disturbed the stability of the surface water cycle due to excessive consumption and pollution. As a result, in the recent past the need for improved surface water treatments became obligatory to provide good quality drinking water to devoid of microbial contamination. However, in the past, the surface water treatments were carried out based on conventional physico-chemical processes including coagulation, flocculation, sand filtration, disinfection, etc. The conventional technologies are used for long time which endow with good quality and portable water, and their design and operation are well understood by the operators. Lately, membrane alternatives have drawn mounting attention because these technologies have highly developed and membrane systems significantly reduce the space requirement to treat a given flow, reduce chemical requirements, and produce water which is easily disinfected and less prone to produce adverse disinfection by-products.

There are different types of membranes available to treat the surface water and they bring several advantages such as effective removal of turbidity, total organic carbon, and micro particles from the surface water. Besides the improvement of prescriptions, development of new concepts and usage of new technologies like nanotechnology have encouraged the use of ceramic membrane. In recent years, ceramic membranes have found to be an attractive alternative to organic membranes, especially for treatment of surface water. In addition, the ceramic membrane has many unique advantages, when compared with the traditional filters and polymer or organic membranes, such as excellent resistance to acid/alkaline and oxidation chemicals, solvent stability, high permeate production at relatively low pressure, high thermal stability, fine separation with narrow pore size distribution, excellent mechanical and abrasive resistance, extremely long working period, high recoveries, hydrophilic membrane surface, and, easy to clean and sanitize with short backwash interval (with air flush). Nevertheless, the membrane fouling is the major drawback of the direct ceramic membrane system. Consequently, it requires appropriate pre-treatment system like chlorination, adsorption by powdered activated carbon (PAC) or coagulation/flocculation to trim down the membrane fouling. Yuasa, et al. (2003) found that when the ceramic membrane system was combined with pre-treatment processes, the filtration achieved high rate of pollutant removal from surface water, where they achieved 99.7% of turbidity removal.

The ceramic membrane is being applied in the drinking water treatment over two decades and there is a great potential for application of the ceramic membrane technology in developing countries. Nevertheless, the operation of this technology depends on the specific characteristics of surface water and other local factors such as temperature, pH, turbidity, etc. Thus, this study was conducted to evaluate the removal efficiency of the natural organic matter and pathogens (bacteria and protozoa) from the surface water using a hybrid ceramic microfiltration (CMF) system, with the combination of pre-treatments such as chlorination, PAC adsorption, and coagulation in the ambient conditions. In addition, the removal of *Giardia* and *Cryptosporidium* was also investigated in this study as both organisms have caused numerous deaths in recent years in USA, Canada and some other countries (AWWA, 1999).

Pasts studies related to drinking water treatment with ceramic microfiltration systems are mainly conducted in developed countries like Japan (Yonekawa et al., 2004; Oh et al., 2007), Germany (Lerch et al., 2005) and Norway (Meyn et al., 2008). The studies were conducted with different but relatively low feed water (dissolved organic carbon) DOC concentrations. Most of the DOC values of feed water were less than 3 mg/L (Yonekawa et al., 2004; Oh et al., 2007; Lerch et al., 2005). However, Meyn et al. (2008) conducted the studies with feed of 5.5 mg/L DOC. It is important to investigate the treatment system behavior under feed conditions with higher DOC concentrations. This study was conducted with feed water of DOC concentration ranging from 6.9 – 10.5 mg/L, which is a relatively higher value than past studies.

2. Materials and methods

2.1 Feed water

The rainwater runoff storage pond water from Asian Institute of Technology (AIT), Thailand was used as a surface water source for this study. The pond water was sent through a raw mesh screen to remove the floating and big particles, and, then stored in a storage tank. Table 1 lists the characteristics of the AIT pond water used in this study.

Table 1: *Characteristics of AIT pond water*

Parameter	Unit	Value
pH	-	6.5 - 8.2
Temperature	°C	26 – 31
Turbidity	NTU	5.18 – 23.1
Conductivity	µs/cm	259 – 505
Micro-particle, 5-15 µm	Count/mL	1,230 – 11,448
TS	mg/L	198 – 315
TSS	mg/L	8 – 21
TOC	mg/L	10.05 – 12.5
DOC	mg/L	6.86 – 10.51
Total Fe	mg/L	0.02-0.09
Total Mn	mg/L	0.07-0.15

2.2 Experimental set-up

A pilot scale experiment was conducted with dead-end mode CMF. The CMF pilot system unit was made in Japan and the process of the pilot system was controlled automatically. The hybrid CMF system was combined with pre-coagulation unit with 2 minutes hydraulic retention time (HRT) to augment the efficiency of the filtration. In addition, the backwashing unit was accompanied to clean the fouling layer inside the channels of the membrane mechanically. The ceramic membrane filter (NGK insulators, Ltd.) with a pore size of 0.1 µm was installed vertically inside a module casing (a stainless steel tube). Furthermore, the 45 cm long ceramic membrane had 55 channels and each channel had inner diameter of 2.5 mm. The membrane was operated at constant flux of 50 $L/m^2.h$ in all the experimental runs. The operational conditions of the CMF are tabulated in the Table 2.

Table 2: *Operational conditions of the CMF system*

Description	Value
Effective membrane area	0.18 m^2
Membrane flux	1.2 $m^3/m^2/day$
Membrane filtration rate	0.2 m^3/day (150 ml/min)
Filtration time (backwash interval)	2 hours
Backwash pressure	500 kPa

Three different scenarios were set-up to achieve the proposed objectives of this study namely, (a) Scenario 1: Direct CMF, (b) Scenario 2: Coagulation and Flocculation + CMF, and (c) Scenario 3: PAC adsorption + Coagulation and Flocculation + CMF. In the scenario 1, the feed water was directly filtered through the ceramic membrane and the performance was compared with the other two scenarios. During the scenario 2, the raw water was pumped into the coagulation tank from the storage and then passed through the flocculation tube. The coagulant poly aluminum chloride (PACl) was added with an optimum dosage of 2 mgAl/L which was determined by the Jar test. In addition, the flocculation process was used to improve the pollutant removal efficiency of the ceramic membrane. The flocculated water was sent to the inlet channels of the ceramic membrane for filtration and the filtrate was collected in the filtrate tank after passing it through the pressurized tank. On the other hand, in the scenario 3 the powder activated carbon (PAC) was added with the dosage of 20 mg/L (the optimum value found through series of Jar tests conducted with PAC doses of 0-250 mg/L) before coagulation-flocculation process to augment the performance of the hybrid system. Further, in all three scenarios, the backwashing unit was automatically activated after every two hours of filtration to reduce the cake fouling in the ceramic membrane. The backwashing unit was operated at 500 kPa using filtrate for 5 seconds which was ameliorated by NaOCl, and air blow down was followed at 200 kPa for 5 seconds. In this study low backwash pressure was maintained as a creative approach to evaluate the fouling potential by lowering the backwash intensity. Besides, ex-situ chemical cleaning with citric acid solution 1% (for 24 hrs) and NaOCl 0.3% (for 24 hrs) was done to remove the irreversible fouling which can not be removed by regular backwashing, when the transmembrane pressure (TMP) reached 100-120 kPa. The overall hybrid CMF process is illustrated in the Fig.1.

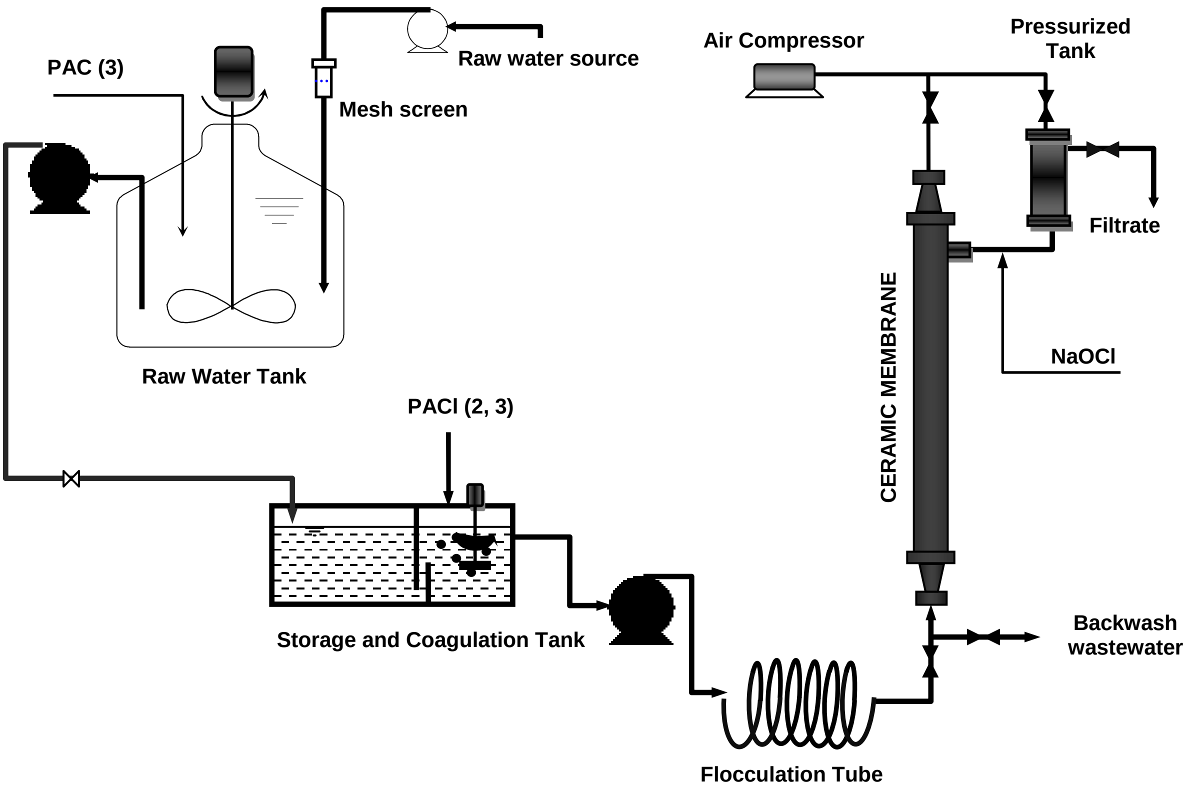

Fig. 1: *Simplified flow diagram of the CMF system*

2.3 Analytical methods

Standard Methods (APHA et al., 1998) were followed to measure the Alkalinity, total solids (TS), total suspended solids (TSS), free chlorine, total coliform and fecal coliform. The total organic carbon (TOC) and the dissolved organic carbon (DOC) were measured using Total Organic Carbon Analyzer (TOC 5000A, Shimadzu, Japan). The DOC is defined as the organic carbon which remains after filtration of the sample through 0.45 μm filter (Jarusuthirak, et al. 2007). In addition, total Fe and total Mn were measured using atomic absorption spectrophotometer (AAS) Z-8230. Conductivity and turbidity were measured with Conductivity Meter (WTW-330i) and Turbidimeter (HACH 2100N) respectively. The measurement of temperature, pH and TMP were noted down daily to monitor the stability of the system. In this study *Giadia* and *Cryptosporidium* were focused as they are the most common protozoa pathogens with the body size of 5 to 15 µm (AWWA, 1999). The particle counter MLC-7P (made in Japan) was used to count the particle numbers of *Giardia* and *Cryptosporidium* (*Protozoa)*. The size range of particles that can be measured by the counter was 1 to 25 µm. The particle counter was initially set-up with tap water passing through 0.45 µm. When the particle count was less than 50 Counts/mL the counter was considered as stabilized. Then the pre-treated samples (pretreatment was done by filtering through a screen with pore size of 53 µm) were injected with a flow rate of 50 mL/min. The readings were taken after allowing system to stabilize, where minimum variation of repeated observations was recorded. The concentrations of particle size ranging from 5 – 15 µm were reported as an indicator for degree of presence of *Giardia* and *Cryptosporidium* (*Protozoa).*

3. Results and discussion

3.1. Comparison of filtration time, TMP & TMP recovery

As mentioned earlier, the three different scenarios were compared to optimize the best scenario to treat the surface water. The Fig. 2 illustrates the change of TMP with the filtration time for different scenarios. The direct CMF had showed the shortest filtration duration of 9 days to reach TMP to 100 kPa. However, with pre-treatment by PACl coagulation-flocculation, the system had showed longer filtration time of 17 days to reach TMP to 100 kPa. Here large molecular compounds and colloidal fraction could be easily flocculated and separated on the microfilter membrane surface. Where as the low molecular size compound with the molecular weight around 1000 Dalton is expected to penetrate into the membrane pores and adsorbed on to the pore walls. Same time major fraction of low molecular weight organics could pass though the membrane. DOC removal efficiencies (section 3.2 and Fig. 4) supports that considerable amount of organics are (40% of DOC) passing through the membrane in the scenario 2. However in scenario 3, another portion of organics were eliminated (only 20% of DOC was passing) through the treatment process. Thus, pre-treatment by adsorption of the low molecular weight compounds using PAC facilitated to prolong the period of treatment further (20 days to reach TMP to 100 kPa) which is the longest filtration period achieved when compared with other two scenarios. Hence, it can be concluded that the pre-treatment processes including coagulation-flocculation and adsorption effectively reduce the membrane foulants in the feed water. As a result the irreversible fouling which can not be effectively removed with the periodical (2 hours) backwash has been reduced. Hence the pretreatment effectively reduce the requirement of chemical cleaning to remove irreversible fouling which leads to longer filtration cycle.

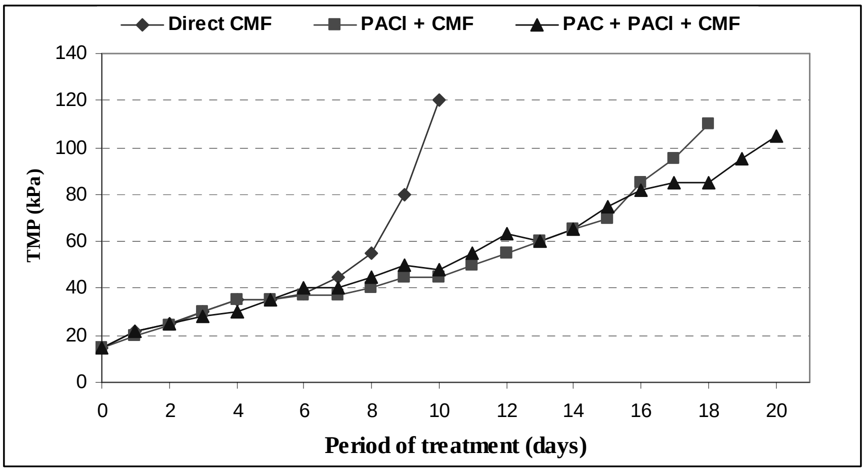

Fig. 2: *Variation of TMP with filtration time*

In addition, the TMP raise in scenario 1 showed rapid growth and low TMP recovery after backwashing. This could be caused by high particle (size less than the membrane pore size) content in the feed water where no pre-treatment was carried out to evade the particle content prior to membrane filtration. In contrast, TMP of the scenario 2 and 3 showed a gentle increment with time (Fig.2). Fig. 3 indicates the TMP recovery after the periodical 2 hour physical backwashing process with 500 kPa pressure (Table 2) during the system operation. The average TMP recovery was higher in the scenario 2 and 3 when compared with scenario 1. The increments of scenario 2 and 3 are 30% and 40% of the scenario 1 respectively. With the pretreatment applications, the part of the fouling components which causes irreversible fouling has been removed and hence the TMP recovery after backwashing process has increased. Study of Yonekawa et al. (2004) with similar type of ceramic membrane obtained 98% of water recovery after backwashing process in a hybrid CMF system as the reversible fouling was removed periodically. In addition, they found that the chemical enhanced backwashing increased the water recovery to 99.2%. Hence, it can be concluded that the reduction of micro particles flow into the ceramic membrane was occurred as a result of pre-treatment process in the hybrid CMF system.

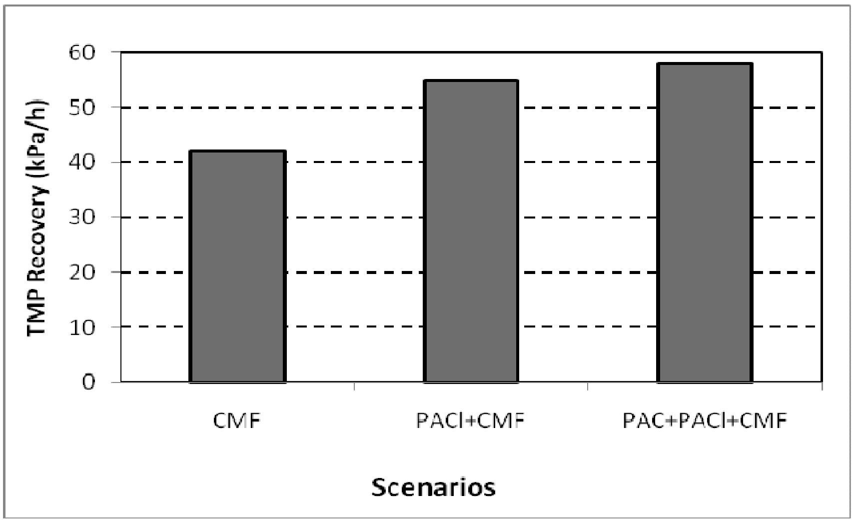

Fig. 3: *TMP recovery after backwashing operation*

Similarly, in this study, on top of backwashing process, the chemical cleaning was done once the TMP reached to 100-120 kPa. As mentioned in section 2.2, the chemical cleaning of a ceramic membrane in this study contains two stages namely, stage 1: cleaning with 1% citric acid and stage 2: cleaning with 0.3% NaOCl. Citric acid was used to dissolve the inorganic matters existing in the form of both inorganic and inorganic-organic complexes like ion-organic compound. During the chemical cleaning stage 1, portion of the TMP values of fouled membranes were recovered. The measured TMP after this step were 60 kPa, 25 kPa and 20 kPa for scenario 1, 2 and 3 respectively. In addition, after stage 2 chemical cleaning the TMP was further reduced to 15 kPa for all three scenarios.

In the scenarios 2 and 3, the feed water was pre-treated by PACl (coagulation-flocculation) alone, or PACl and PAC (adsorption), and as a result significant amount of dissolved organics and colloids were formed into flocs or the dissolved organics were adsorbed by the PAC particles. According to Suzuki & Chihara (1988) during PAC pretreatment process, dissolved organics were removed by adsorption, and suspended solid organics were by heterogeneous coagulation, at the same time. Hence untimely the dissolved organics and colloids were fixed to the larger flocs which can be removed by microfiltration through surface filtration. Accordingly, these flocs were removed effectively from the membrane surface by backwashing process (Zhao et al., 2005). In addition, as cited earlier the ions and colloidal fouling were also brought down due to pre-treatment process in the system. Thus, it can be concluded that the chemical cleaning by citric acid and the pre-treatment process in the hybrid CMF system can increase the TMP recovery. Further, it is noted that the scenario 2 and 3 are the better treatment sequences when compared with the scenario 1 as the pre-treatment processes reduces the chemical cleaning load.

3.2. Pollutant removal of the system

The total coliform, fecal coliform, and TSS were eliminated completely by the system in the case of all three scenarios during the steady state operation (Fig. 4). With the pore size of 0.1 μm, the ceramic membrane was able to remove the bacteria and TSS effectively which is one of the attractive benefits of the CMF. Moreover, the addition of NaOCl into the filtrate tank for enhancing the backwashing process contributed for successful disinfection of remaining bacteria in the filtrate. In addition, the Fig. 4 explains the key roles of the pre-treatment processes based on pollutant removal efficiency. The turbidity removal through scenarios 1, 2 and 3 were more than 99.3%. Other than the above mentioned pollutants, around 25%, 60% and 80% of both TOC and DOC were eliminated through scenarios 1, 2, and 3 respectively. This was due to the removal of both low and high molecular weight organic compounds through physico-chemical treatment of coagulation-flocculation and adsorption processes. Another study conducted with a pilot-scale PAC-MF system had been functioning for one year without withdrawal and replacement of PAC with an average DOC removal rate of around 80% (Kim et al., 2006; Oh et al., 2007). Hence, it confirms that the pre-treatment process can increase the removal efficiencies of the system. The total Fe removal through scenarios 1, 2 and 3 were 92%, 98% and 96% respectively (Fig. 4). In the case of removal of *Giardia* and *Cryptosporidium*, the hybrid CMF system has removed micro particles between 5 – 15 µm up to the undetectable level for all three scenarios. Since maximum reported raw water particle count is 11,448 Count/mL (Table 1), the system has shown over log-3 removal. Where the size of *Giardia* and *Cryptosporidium* (5 – 15 µm) are much grater then the nominal pore size of membrane (0.1 µm), even higher degree removals are expected. Studies of Lerch et al. (2005)

with same type of ceramic membrane (0.1 µm; NGK insulators, Ltd.) has shown over log-4 removal, where the raw water micro particle concentration is much higher than this study.

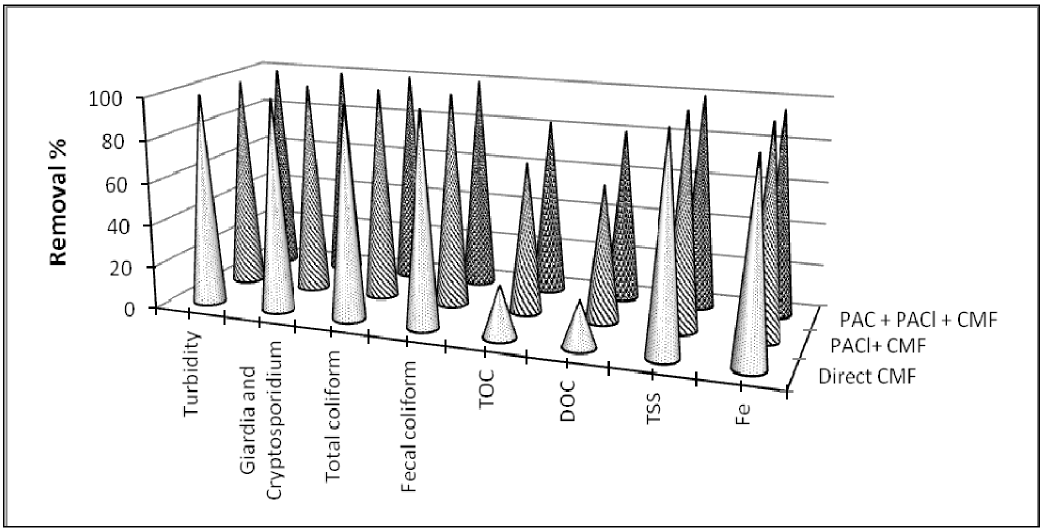

Fig. 4: *Removal rate of major pollutants for different scenarios*

The objective of any treatment technology is to reduce the disease causing organisms and related health hazards to a tolerable or safe level. The level of acceptability varies from country to country and depends on several factors such as environmental, human health and economic conditions. The overall results revealed that the treated water from the system with scenario 3 can be used for drinking purpose as the quality of the water meet the WHO standard requirement. However, the other two scenarios can also be utilized for drinking purpose after further treating for *Giardia* and *Cryptosporidium.*

4. Conclusion

In this study, the key role of pre-treatment processes combined with the pilot scale CMF system on surface water treatment was investigated with different combination of pre-treatment scenarios. It was found out that the pollutants removal efficiencies and performance of the hybrid CMF system differ depending on the pre-treatment process and operational conditions. In conclusion, the effectiveness of ceramic membrane filtration can be enhanced by pretreatment such as coagulation, PAC as well as by chemical backwashing.

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