Biofilter pretreatment for the control of microfiltration membrane fouling

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Abstract
A pilot scale biofilter pretreatment - microfiltration system (BF-MF) was operated to investigate the effect of biofilter treatment in fouling reduction of microfiltration. Biofiltration was expected to reduce the membrane fouling by removal of turbidity and metal oxides. The hollow-fiber MF module with a nominal pore size of 0.1 μm and a surface area of $8m^2$ was submerged in a filtration tank and microfiltration was operated at a constant flux of 0.5 m/d. Biofiltration using polypropylene pellets was performed at a high filtration velocity of 320 m/d. Two experimental setups composed of MF and BF/MF, i.e., without and with biofilter pretreatment, were compared. Throughout the experimental period of 9 months, biofilter pretreatment was effective to reduce the membrane fouling, which was proved by the result of time variations of trans-membrane pressure and backwash conditions. The turbidity removal rate by biofiltration varied between 40% to 80% due to the periodic washing for biofilter contactor and raw water turbidity. In addition to turbidity, metals, especially Mn, Fe and Al were removed effectively with average removal rates of 89.2%, 67.8% and 64.9%, respectively. Further analysis of foulants on the used membranes revealed that turbidity and metal removal by biofiltration was the major effect of biofiltration pretreatment against microfiltration fouling.

Keywords
Biofilter pretreatment; manganese; membrane fouling; metals; microfiltration

INTRODUCTION

Irreversible membrane fouling is one of the major limiting factors in the use of membrane technology for water treatment since it results in a drop of filtration flux and the increase of physical/chemical cleaning frequency.

In spite of a lot of information, it is difficult to make an estimation of the fouling mechanism and extent because of its complexity depending upon raw water quality. For example, particulate fouling, organic and inorganic fouling, and biofouling can be caused by various impurities in raw water. The foulants could be particulate matter such as suspended or colloidal solids (clay, silica, metal oxides, etc.), organic matter such as natural organic matter, scaling materials such as carbonates, sulfates and silicates, and microorganisms and their metabolism by-products.

Thus, the introduction of pretreatment processes is very important not only to obtain desired water quality but also to achieve the improvement of membrane performance. In accordance with each membrane process and raw water quality, various strategies are available for reducing membrane fouling. For instance, physical membrane cleaning with periodic backwash and/or air scrubbing, or cross-flow filtration (Takizawa et al., 2000a) has been applied at many plants around the world. In addition to those physical cleaning, pretreatment processes such as pre-chlorination (Huang et al., 2000), UV irradiation (Takizawa et al., 1995; Ohtaki et al., 1998; Huang et al., 2000) and ozonation (Takizawa et al., 1996) have been evaluated extensively and found to be effective to reduce fouling by prevention of microbial growth.

Chlorination is an effective method for fouling control but it sometimes causes progressive membrane fouling (Babcl et al., 2000). One of problems with the chlorination is inorganic fouling caused by facilitating oxidation of manganese and iron, which may become a serious problem in ground water containing a high level of manganese and iron. Takizawa et al. (2000b) have evaluated the effect of biofilter pretreatment for the ground water on reduction of manganese fouling.

In this study, biofilter treatment was used as a pretreatment process followed by microfiltration. Biofiltration makes use of the activity of biofilms attached on filter media and is expected to reduce some fouling, especially fouling due to particulate matter.

Our main objectives in this study were to evaluate the effect of biofilter pretreatment of raw water with low manganese concentration and to elucidate the fouling mechanisms through the investigation of foulants.

EXPERIMENTAL METHOD

The study was conducted using a pilot-scale of biofilter pretreatment and microfiltration systems. Experimental setup was located in Tamagawa Water Treatment Plant that treats Tama River water. Two series of processes, with (BF/MF process) and without (MF process) biofilter pretreatment, were operated simultaneously and the results were compared (Figure 1). The specifications of pilot plant and the operating conditions are shown in Table 1.

The hydrophilic polyethylene hollow fiber membranes were used and had a nominal pore size of 0.1 μm and an effective surface area of 8.0 m^2. The membrane module was submerged in filtration tank equipped with pneumatic scrubbing and hydraulic backwash system for periodic physical cleaning. Microfiltration was operated in a dead-end vacuum filtration mode with constant operating flux of 0.5 m/d and one filtration cycle consisted of 30 minutes of filtration and 1 minute of cleaning. For prevention of biofouling, sodium hypochlorite solution (7-8 mg/l as residual chlorine) was added to both membrane filtrates that were used as backwash water. In addition, 7-8 liter of the filtration concentrates was drained out once every four filtration cycles.

The fixed bed biofilter contactor was packed with the tubular polypropylene media (I.D. 3 mm, O.D. 4 mm, and L 5 mm) and down-flow filtration was operated at a high filtration velocity of 320 m/d. Biofilter media were cleaned once a week by raw water flushing and air scrubbing.
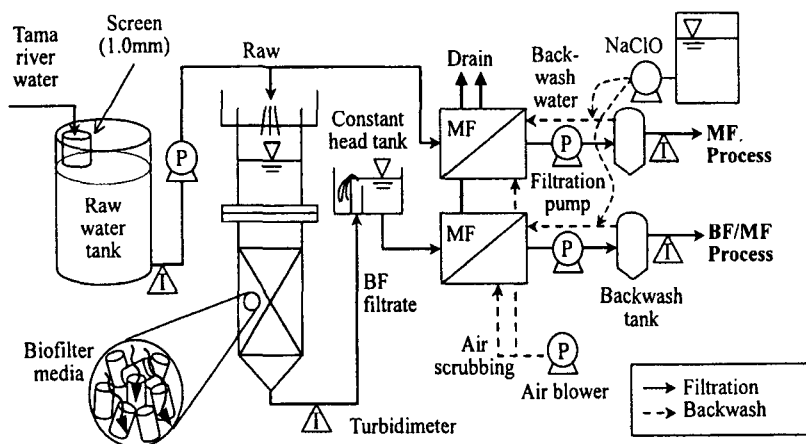

Figure 1. Schematic diagram of BF-MF system composed of two process, BF/MF and MF

Table 1. Specification of biofilter pretreatment and microfiltration system

Microfiltration	Membrane type	Polyethylene hollow fiber microfilter (hydrophilic) Nominal pore size: 0.1 μm, Surface area: 8.0 m^2
	Filtration method	Dead-end, constant flux (0.5 m/d)
	Filtration cycle	Filtration (30 min) - Physical cleaning (1min)
	Physical cleaning	Backwash for 20 sec during 1 min of air scrubbing NaClO addition to backwash water (7-8 ppm) Drain once every four cycle (7-8 L)
Biofilter pretreatment	Contactor type	Fixed bed, Down flow (φ 20 mm×2,400 mm H)
	Filter media	Coarse polypropylene tube (5mm L×4mm o.d.×3mm i.d.)
	Packing depth	1,200 mm
	Filtration rate	320 m^3/m^2/d (EBCT 6 min)
	Filter cleaning	Once a week with air scrubbing and flushing by raw water

Trans-membrane pressure (TMP), turbidity, and temperature data were collected in every one hour through the telephone line and stored in a computer at the University of Tokyo. Since turbidimeters (1720C/D, HACH) were calibrated by standard kaolin solution, turbidity was recorded as kaolin unit that was equivalent to mg kaolin/liter.

Each sample of process steps was collected once a week and analyzed for several water quality parameters including DOC, UV absorbance, total and dissolved metals, and NH3-N, etc. Samples for analysis of DOC, UV absorbance and dissolved metal concentrations were filtrated by 0.1μm microfilter (hydrophilic Durapore, Millipore). Total and dissolved organic carbon concentrations were analyzed by TOC analyzer (TOC-5000, Shimadzu) and metal concentrations were analyzed by ICP-AES (Optima 3000DV, Perkin Elmer). UV absorbance at 260 nm with U-2010 spectrometer (Hitachi) and NH3-N with DR/2010 spectrometer (HACH) were also analyzed.

RESULTS AND DISCUSSIONS

The average water qualities

The pilot plant experiments were carried out for about 9 months from April to December 2000. The average water quality for the entire experimental period is shown in Table 2.

For pH, temperature and DO concentration there was no significant difference between raw water and biofiltrated water. Temperature was varied from 10 to 31°C. On the average, 61.5% of ammonia removal by biofilter was observed under the very low concentrations in raw water. On the other hand, biofilter had little effect in removal of DOC and UV260 with the removal rates of 1.5 % and 1.1 %, respectively. The average turbidity of raw water and biofiltrated water were 2.22 units and 0.98 units. The turbidity removal rate by biofiltration was 55.9 % in average and varied from 40 % to 80 % depending on biofilter cleaning (Figure 2).

Inflow of high turbidity over 20 mg/l caused a temporary increase of trans-membrane pressure, particularly in MF process. While TMP was recovered after decrease of turbidity, turbidity removal by biofilter probably contribute to reduction of the particulate loading to the microfilter.

Table 2. The average water qualities for raw water and biofiltrated water   ( ): Removal rate (%)

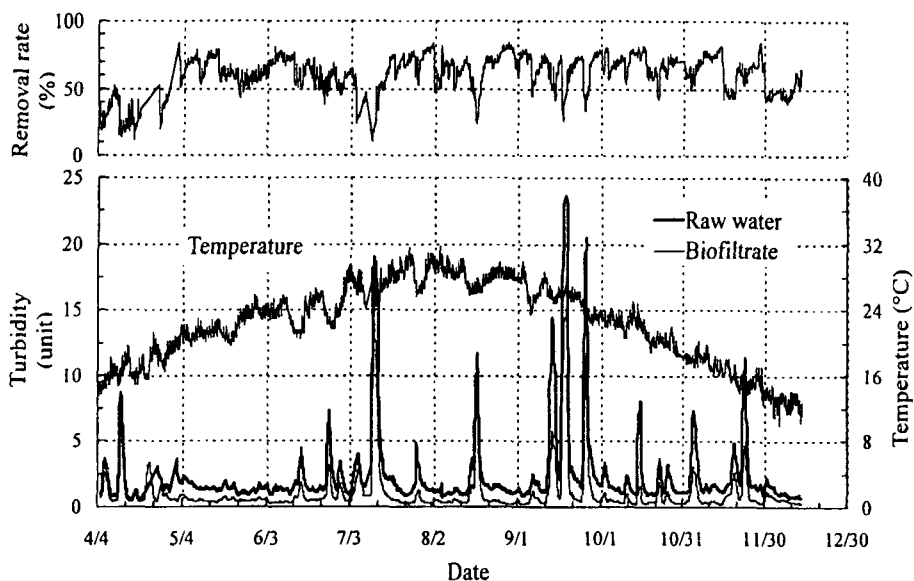| Sample	pH	Temp. (°C)	DO (mg/L)	Turb. (unit)	NH3-N (mg/L)	DOC (mg/L)	E260 (1/cm)
Raw water	7.54	21.3	7.33	2.22	0.13	2.05	0.181
Biofiltrated water	7.56	21.7	7.26 (1.0)	0.98 (55.9)	0.05 (61.5)	2.02 (1.5)	0.179 (1.1)

Figure 2. Turbidity removal by biofiltration

The time course of filtration resistances and backwash conditions

The membrane resistances measured by pure water before the use were 5.85×10^{11} m^{-1} and 6.05×10^{11} m^{-1} for BF/MF and MF processes, respectively, which corresponded to the filtration pressures of 4.0 kPa and 4.1 kPa at a constant flux of 0.5 m/d. Both membranes were then compared for their long time performance with raw water and biofiltrated water.

As shown in Figure 3, the BF/MF process exhibited the stable filtration keeping on less increase of filtration resistances than MF process. While the microfiltration of MF process was stopped in about 9 months due to severe membrane fouling, BF/MF process remained stable at TMP lower than 28 kPa. Consequently, biofilter pretreatment were proved to be effective to reduce membrane fouling.

One week after starting filtration, dramatic increments in resistances were observed for both processes. They disappeared gradually one week later when the addition of chlorine to backwash water was started. It was due to the elimination of irreversible biofouling by prevention of microbial growth on membrane surface. Sodium hypochlorite solution is known to be an effective cleaning agent for some biological and organic foulants. Thus, the progress of membrane fouling was considered to be caused by other reasons than biofouling.

After controlling the biofouling by chemical agent, filtration resistances for both processes were stable for about 6 months and followed by significant increment in MF process during last 3 months. The increase of filtration resistance in MF process can be explained by the change of backwash resistance as well as possible change of raw water quality. Since backwash was conducted at a initial pressure of 0.2 MPa in backwash tank and backwash time was limited to 20 seconds, it resulted in increase of backwash resistance with increase in the filtration fouling.

As shown in Figure 4, the fact that backwash volume decreased with time was considered to explain the rapid increase of filtration resistance in the latter experimental period. Backwash resistance of MF process was always higher than that of BF/MF process and increased rapidly. In was expected that decrease of backwash volume resulted in a drop in backwash efficiency and might cause more rapid increase of filtration resistance.
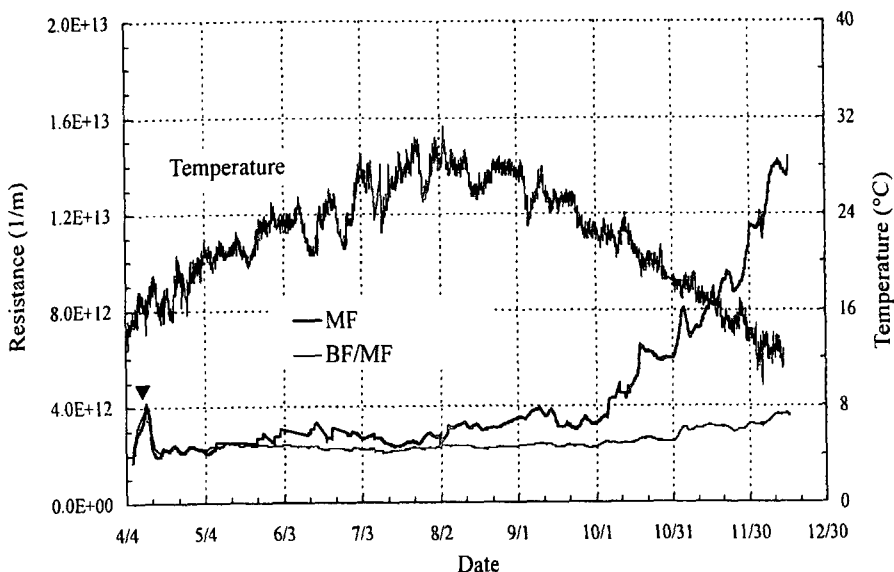
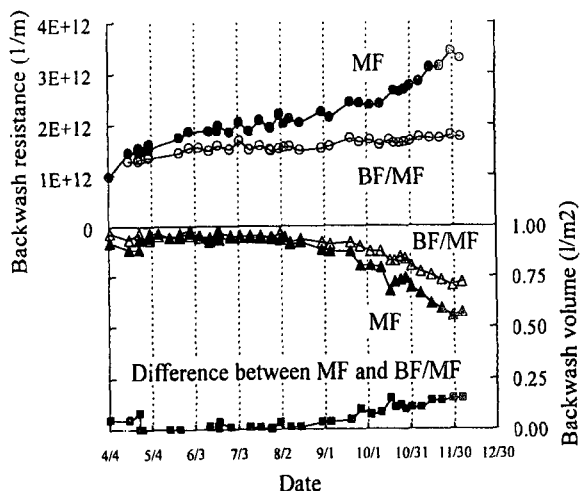**Figure 3.** The time course of filtration resistance and temperature

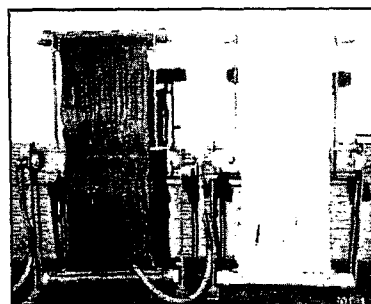**Figure 4.** The time course of backwash conditions

Photo 1. Membrane modules at the end of experiment (left: MF process with brown color, right: BF/MF process with white color)

During the same experimental period, brown color was observed on the membrane used in MF process and foulants were strongly attached on the membrane surface, while BF/MF process did not show any significant change in color (Photo 1). From the color formed on the membrane surface, it was thought that membrane foulants were deposits of iron and manganese oxides.

Metal removal by biofiltration and microfiltration
The average metal concentrations at each process step are shown in Table 3. It was found that biofilter had higher removal effect for Al, Fe, and Mn than other metals such as Ca, Na, Mg, K, and Si In order to estimate the amount of metals accumulated on membrane surface, mass balance for Al, Fe and Mn were calculated based on the average metal concentrations (Figure 5).

Table 3. The average metal concentrations at each process step ( ): Removal rate (%)

Metal	Unit	Raw water	BF filtrate		Filtrate				Concentrate	
					MF		BF/MF		MF	BF/MF
Ca		21.57	21.52	(0.2)	21.55	(0.1)*	21.68	(-0.7)**	23.73	22.25
Na		10.52	10.51	(0.1)	10.88	(-3.4)	10.73	(-2.1)	10.39	11.07
Mg		4.71	4.68	(0.6)	4.67	(0.8)	4.62	(1.3)	5.39	5.02
K	mg/L	3.22	3.22	(0.0)	3.21	(0.3)	3.23	(-0.3)	3.44	3.39
Si		8.06	7.84	(2.7)	7.78	(3.5)	7.76	(1.0)	11.76	8.90
Al		0.28	0.10	(64)	0.03	(89)	0.03	(70)	3.61	0.85
Fe		0.32	0.10	(69)	0.05	(84)	0.05	(50)	4.54	1.28
Mn	µg/L	34.2	3.7	(89)	2.1	(94)	2.0	(46)	545.2	66.6
Cu		7.8	6.7	(14)	6.3	(19)	6.5	(3)	24.9	13.0

* Removal rate for raw water, ** Removal rate for BF filtrate

In MF process (Figure 5 (a)), the ratios of Al, Fe and Mn attached on the membrane were 60.9 %, 53.3 % and 58.1 % for raw water, respectively. On the other hand, in BF/MF process (Figure 5 (b)), the ratios of Al, Fe and Mn attached on the membrane decreased to 17.8 %, 6.2 % and 0.7 %, respectively since these metals were largely removed by biofilter pretreatment. Thus it was assumed that these metals contributed to membrane fouling and metal removal by biofilter was one of the main effects for membrane fouling reduction in BF/MF process.

Organic matter, which was hardly removed by biofilter in this study, can also be considered as an important membrane foulant. Takahashi et al. (1994) have examined the organic adsorption to membrane using coliphage Qβ as model organic matter and the same type of membrane as in this study. Their results indicated that coliphage Qβ was adsorbed in a very short time and did not exceed the some adsorption levels. Kim et al. (2000) have carried out filtration experiments using 8 kinds of model organic matter such as humic acid and tannic acid, etc. They observed that fouling rate by adsorption of these model organic matter increased in proportion with increase in filtration volume but corresponding filtration resistance were so small that organic adsorption were not the main cause of membrane fouling.
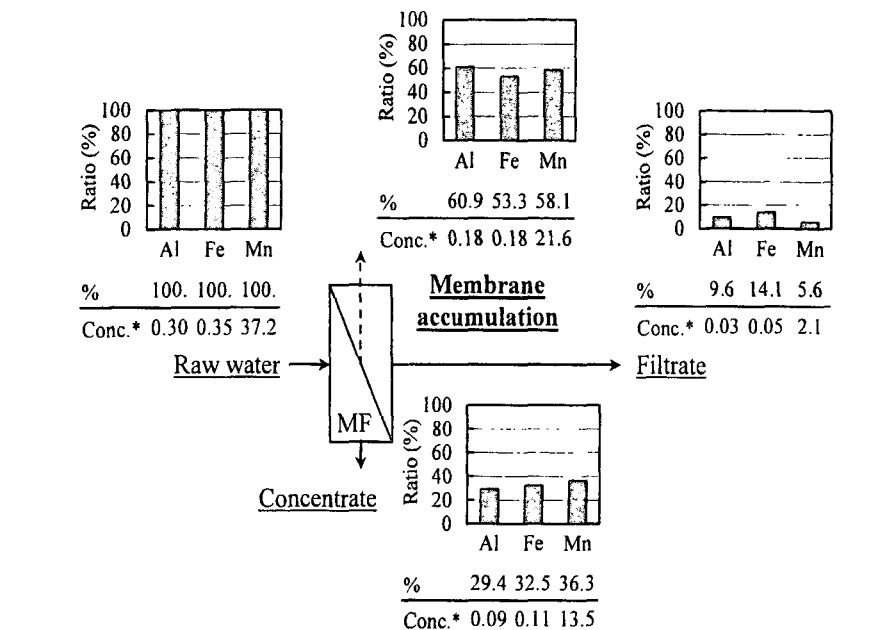
With these results and the aforementioned review, it was supposed that organic matter was not the primary cause of membrane fouling in this study. But there is still possibility that organic property changed by biofilter can affect membrane fouling and can be closely related to metal accumulation. Therefore, in spite of very little organic removal by BF, further study is needed to investigate the role of organic matters for membrane fouling.
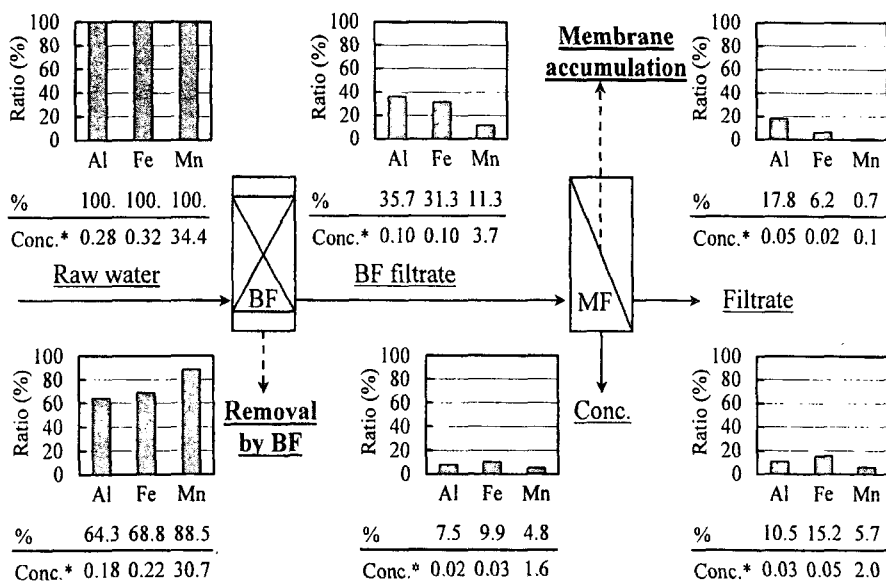

CONCLUSIONS

Biofilter pretreatment using polypropylene media at high filtration velocity proved to be effective for the control of microfilter fouling. The biofilter pretreatment effect for the control of membrane fouling was observed by time course of filtration resistance and backwash conditions. A rapid increase of filtration resistance for MF process could be caused by decrease in backwash volume as well as possibly the change of feed water quality.

It was assumed that the enhancement of filtration performance of BF/MF process was obtained by the change of water quality by biofilter treatment. Turbidity removal rate was 55% on the average and varied from 40% to 80% due to biofilter cleaning and inflow of high turbidity. Some metals, especially Al, Fe and Mn, were effectively removed by biofilter and also by microfilter. The removal rates for Al, Fe and Mn by the biofilter were 64 %, 69 % and 89 %, respectively. From the mass balance for these three metals, the ratios of membrane accumulation of Al, Fe and Mn were 60.9 %, 53.3 % and 58.1 % for MF process while 17.8 %, 6.2 % and 0.7 % were obtained for BF/MF process, respectively. Hence the removal of turbidity, Mn, Fe, and Al is considered to be the major effect of the biofilter for the control of membrane fouling.

In spite of very low removal rate for DOC (1.5%) and UV absorbance (1.1 %) by biofilter, further analysis would be necessary to identify the role of organic matter which can serve as organic foulants and be related to metal accumulation.

Membrane accumulation

	Al	Fe	Mn
%	60.9	53.3	58.1
Conc.*	0.18	0.18	21.6

	Al	Fe	Mn
%	100.	100.	100.
Conc.*	0.30	0.35	37.2

Raw water → MF → Filtrate

	Al	Fe	Mn
%	9.6	14.1	5.6
Conc.*	0.03	0.05	2.1

Concentrate

	Al	Fe	Mn
%	29.4	32.5	36.3
Conc.*	0.09	0.11	13.5

(a) MF process

	Al	Fe	Mn
%	100.	100.	100.
Conc.*	0.28	0.32	34.4

Raw water

	Al	Fe	Mn
%	35.7	31.3	11.3
Conc.*	0.10	0.10	3.7

BF filtrate

Membrane accumulation

	Al	Fe	Mn
%	17.8	6.2	0.7
Conc.*	0.05	0.02	0.1

→ BF → MF → Filtrate

Removal by BF

	Al	Fe	Mn
%	64.3	68.8	88.5
Conc.*	0.18	0.22	30.7

Conc.

	Al	Fe	Mn
%	7.5	9.9	4.8
Conc.*	0.02	0.03	1.6

	Al	Fe	Mn
%	10.5	15.2	5.7
Conc.*	0.03	0.05	2.0

(b) BF/MF process

Figure 5. Mass balance for total Al, Fe and Mn. **(a)** MF process, **(b)** BF/MF process
(* Concentration units: g/m^3 for Al and Fe, mg/m^3 for Mn)

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