

## The effect of particulate material and the loading of bacteria on a high dose PAC-MF system

M.M.T. Khan\*, H.-S. Kim\*\*, H. Katayama\*, S. Takizawa\* and S. Ohgaki\*

\* Department of Urban Engineering, The University of Tokyo, Tokyo 113-8656, Japan

\*\* Department of Water Supply Engineering, N.I.P.H., Tokyo 108-8638, Japan

(E-mails: [taimur@env.t.u-tokyo.ac.jp](mailto:taimur@env.t.u-tokyo.ac.jp); [kimhs210@iph.go.jp](mailto:kimhs210@iph.go.jp); [katayama@env.t.u-tokyo.ac.jp](mailto:katayama@env.t.u-tokyo.ac.jp); [takizawa@env.t.u-tokyo.ac.jp](mailto:takizawa@env.t.u-tokyo.ac.jp); [ohgaki@env.t.u-tokyo.ac.jp](mailto:ohgaki@env.t.u-tokyo.ac.jp))

**Abstract** The effects of particulate material, their size distribution and the loading of total bacteria based on the age distribution of powdered activated carbon (PAC) were investigated in a hybrid membrane process consisting of an immersed microfiltration membrane system in a completely aerated reactor containing high concentration (40 g/L) of PAC. The laboratory-scale experiments on PAC-MF system were carried out using actual surface water (Tama River, Tokyo), before and after the treatment by a biofilter media. As a result of continuous filtration and backwash experiments, improvement of filtrate water quality and controls of membrane fouling were demonstrated by the addition of PAC. 52% of the PAC fell into a particle size between 22  $\mu\text{m}$  and 45  $\mu\text{m}$ . PAC larger than 125  $\mu\text{m}$  and smaller than 22  $\mu\text{m}$  were 6% and 4% respectively. Due to continuous aeration and Brownian motion of the particles, the size of the PAC inside the reactor was becoming smaller day by day. After 162 days of operation, the particle size from 1–10  $\mu\text{m}$  was higher, which is the suitable size range for the adsorption of bacteria. At the same time, the number of total bacteria was also increasing inside the reactors. The membranes were fouled shortly during end of the operation due to the heavy load of fine particles and high accumulation of bacteria. However, PAC acts as an adsorbent media inside the reactors that causes successive accumulation of total bacteria.

**Keywords** Adsorption; bacteria; membrane fouling; microfiltration; particulate material; powdered activated carbon

### Introduction

With the scarcity of water supply sources, many new water quality parameters come under consideration during purification. These more stringent regulations lead to the conventional processes being replaced by advanced ones (Pontius, 1990). Increasingly stringent regulations related to the treated water quality drive a worldwide interest in membrane technology applications whether for particles or dissolved matter removal (Lebeau *et al.*, 1998).

The term “particulate material” is commonly used to describe the size fraction of aquatic particles larger than 1  $\mu\text{m}$  (Lartiges *et al.*, 2001). The role of this particulate phase in the transport of both organic and inorganic contaminants in a fluvial system has been somewhat overlooked. However, various experimental results suggest that these large suspended particles play a key role in determining the fate of fluvial contaminants, such as heavy metals, microorganisms, and pesticides (Lick and Rapaka, 1996; Leppard *et al.*, 1998). It has been established that particulate material occurs mainly as aggregates of much smaller particles (Zabawa, 1978; Droppo and Ongley, 1992). As a consequence, the total surface area of particulate matter, and hence its capacity to sorb pollutants, is much higher than previously estimated (Perret *et al.*, 1994). The uptake rate of microorganisms by these particulate matters depends on the system itself. However, this kinetics can be identified by any model experiment, even in real situation the result can fluctuate from the reality. But, the basic concept is the same.

Adsorption by powdered activated carbon (PAC) and membrane are a promising unit

process for advanced water treatment. PAC is an important tool for maintaining the safety and aesthetic quality of drinking water. Microfiltration (MF), as well as ultra-filtration (UF), are viewed as particle removal process (Lebeau *et al.*, 1998). However, the membrane process has a limitation that it cannot remove contaminants smaller than its pore size. MF can be combined with PAC adsorption to treat water contaminated with organic compounds and microorganisms (Seo *et al.*, 1997). Once a membrane is fouled, it must be either chemically cleansed or replaced by a new membrane. Only the higher-pressure membrane processes, such as nanofiltration (NF) and reverse osmosis (RO), can remove dissolved organic matter, such as humic substances and taste and odor producing compounds, as well as synthetic organic compounds, such as the trihalomethanes (THMs), to a significant degree. But what if these organic materials could be associated with a particulate phase, for example, with PAC? It should then be possible to use the lower-pressure (and lower operating cost) membrane processes such as MF and UF to remove both natural particulate matter, PAC, as well as the organics adsorbed on the PAC (Clark and Heneghan, 1991; Clark, 1992; Graham, 2000).

On the other hand, a high concentration of PAC can make a cake layer on the membrane surface, thus reducing the effectiveness of physical cleaning by aeration and increasing the filtration resistance. This adverse effect, however, was comparatively less significant than the positive effect of PAC addition. The objective of this paper was to identify the reasons of membrane fouling due to change of particle size, number of bacteria and their effect on transmembrane pressure of the membranes as well as membrane fouling when MF membrane reactors combined with high concentration of PAC were used.

## Materials and methods

### Experimental design and operational conditions of the reactors

Three MF membrane modules were submerged into three 5 L reactors. The specification of the membrane modules and reactors are shown in Tables 1 and 2. Among these three reactors, the input water for Reactor 1 and Reactor 2 was river water (Tama River, Tokyo) without any pretreatment. But, in the case of Reactor 3, the input water (river water) was pretreated by a biofilter (B.F.) reactor (containing polypropylene material). Reactor 1 did not contain any PAC, and was used as a control reactor. Reactors 2 and 3 each contained 40 g/L PAC. A continuous filtration process was carried out to improve the filtrate water quality and control of membrane fouling by the addition of PAC. The particle size distribution of PAC was determined by sieve analysis. The virgin PAC (Product name: Shirotsagi-C of Takeda Chemical Co.; Material: Coconuts shell; Type: JWVA K 113 – 1985) was used during this experiment.

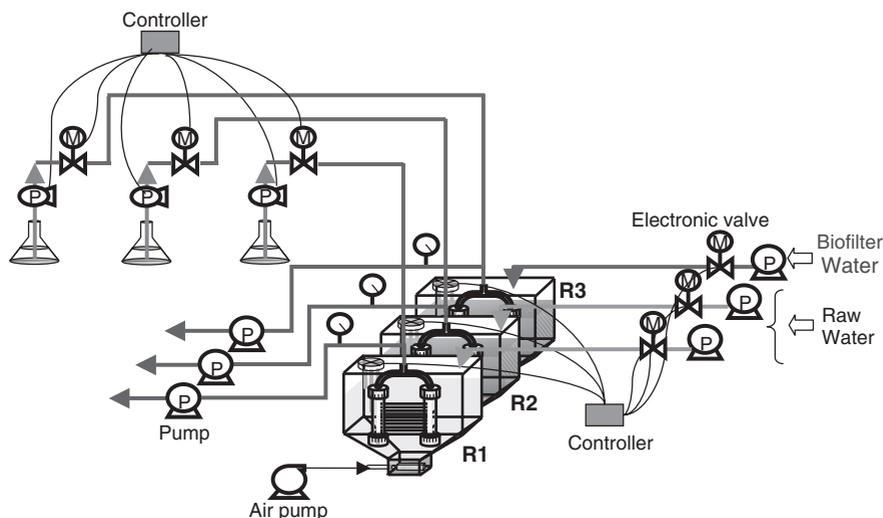
The PAC-MF system is a hybrid membrane process coupled with powdered activated carbon. The schematic flow diagram of the experimental set up is shown in Figure 1. During the filtration experiment, the membrane flux, aeration rate and flow rate of feed pump were constantly maintained at 0.50 m/day, 5 L/min and 25 L/day, respectively. The transmembrane pressure of the membranes was automatically loaded in a data logger and

**Table 1** Characteristics of the membrane module

Material	Hydrophilic polyethylene
Nominal pore size	0.1 $\mu\text{m}$
Outer diameter	0.41 mm
Inner diameter	0.27 mm
No. of fibers	320 (16 $\times$ 20)
Length of fibers	120 mm
Surface area	0.05 m <sup>2</sup>

**Table 2** Characteristics of the reactor

Material	Polyvinylchloride
Thickness	5 mm
Volume	5 L



**Figure 1** Schematic flow diagram of PAC-MF system

was maintained by a computer. At the same time, the suction and backwash of the membranes were controlled by timers with the help of water valves. After every 20 minutes of suction, the duration of backwash was for 2 minutes. When the suction pressure of each membrane was raised to near 50 kPa, that membrane module was physically cleaned for the next period of filtration. In this experiment, all membrane modules were used membranes, but their conditions were quite good. The main purpose was to check whether it is possible to use the used membranes when PAC was virgin. Even many researchers found that PAC with appropriate surface treatment had much better adsorption quality, but this paper did not consider that aspect.

#### Analytical methods

This paper shows only a few selective parameters for the target items. The Coulter Epics Altra Flow Cytometer machine (PN 4237467A, Coulter Corporation, Miami, USA) was used to count the particulate matter, size ranging from 1–20  $\mu\text{m}$ , inside the reactors and also the number of total bacteria. Tables 3 and 4 show the materials which were used to make different protocols for particle and bacteria counting, respectively.

Depending on the type of the samples, totally different protocols were developed for the measurement of total bacteria. This paper will also give brief information related to the suspended solid inside the reactors and two sources of input water, e.g. river water and biofilter

**Table 3** The material description of standard reference particles (SRP) to make different protocols for particle counting

	SRP 1	SRP 2	SRP 3	SRP 4
Sample name	N4 size control L1000	Flow-set fluorospheres	Flow-check fluorospheres	CC size standard L20
Mean size	1088.6 $\pm$ 65.3 nm	3.6 $\mu\text{m}$	10 $\mu\text{m}$	20.09 $\mu\text{m}$
Type of material	Nominal 1000 nm polystyrene latex	A suspension of fluorospheres (fluorescent microspheres)	Type of material is an assayed suspension of fluorospheres (fluorescent microspheres)	Nominal 20 $\mu\text{m}$ polystyrene latex

**Table 4** Type of standard reference particle (SRP) and dye to make protocols for bacteria counting

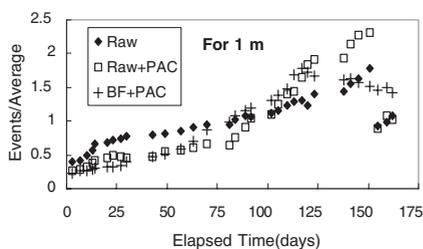
	SRP	Dye
Sample name	Flow-count fluorospheres PN 7507992-E R 2-99	Live/dead bacLight bacterial viability kits L-7007
Mean size	10 $\mu$ m	
Type of material	Polystyrene fluorospheres in an aqueous suspension medium containing a surfactant and 1% formaldehyde	SYTO 9 (the excitation/emission maxima – 480 nm/500 nm)

treated river water. The suspended solid (SS) of these samples was measured by glass fiber filter of 1  $\mu$ m pore size and 47 mm diameter following standard method of analysis.

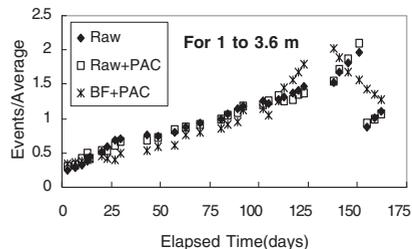
## Results and discussion

The operation of the reactors was started from 3 June 2001 until the present date. This paper shows the data of the specific results of 162 days. As these laboratory-scale reactors were connected to the natural water supply system, the results show real response to the weather fluctuation. Figures 2 to 8 show the particle size distribution inside the reactors.

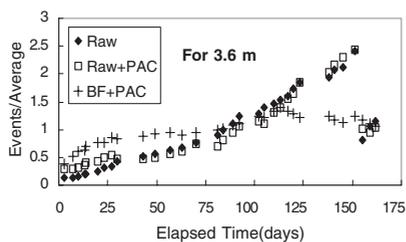
After 151 days of operation, the samples inside Reactor 1 (raw water) and Reactor 2 (raw water + PAC) were changed due to high accumulation of suspended solids (SS) and



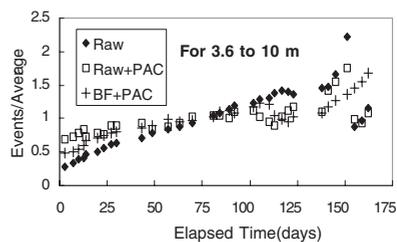
**Figure 2** Particle size distribution of 1  $\mu$ m size in the reactors



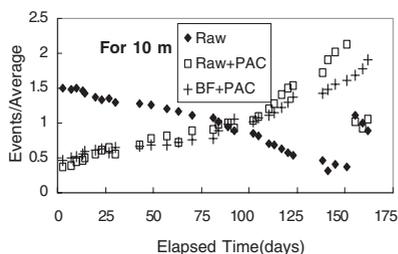
**Figure 3** Particle size distribution of 1–3.6  $\mu$ m sizes in the reactors



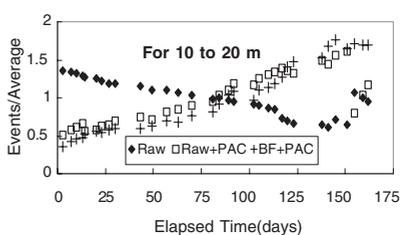
**Figure 4** Particle size distribution of 3.6  $\mu$ m size in the reactors



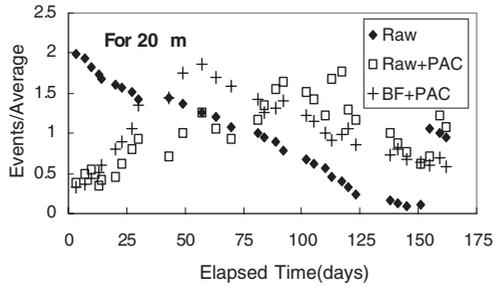
**Figure 5** Particle size distribution of 3.6–10  $\mu$ m sizes in the reactors



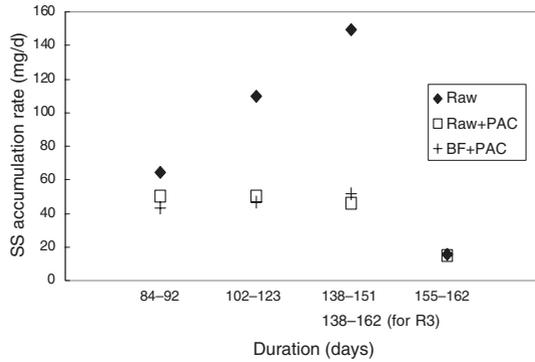
**Figure 6** Particle size distribution of 10  $\mu$ m size in the reactors



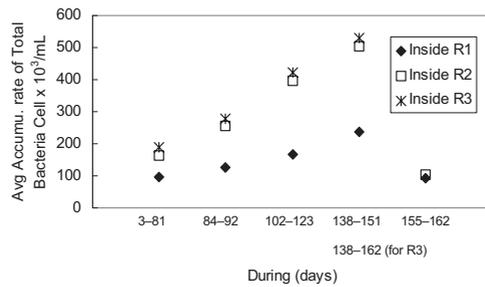
**Figure 7** Particle size distribution of 10–20  $\mu$ m sizes in the reactors



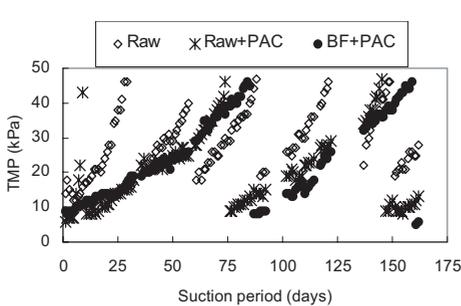
**Figure 8** Particle size distribution of 20  $\mu\text{m}$  size in the reactors



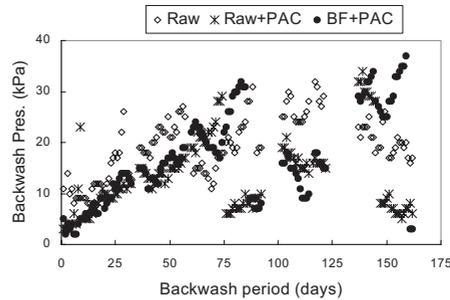
**Figure 9** SS accumulation rate inside the reactors



**Figure 10** Average accumulation rate of total bacteria cell inside the reactors



**Figure 11** Transmembrane pressure of the membrane modules inside the reactors



**Figure 12** Backwash pressure of the membrane modules inside the reactors

shortening of the membrane filter run-time. But, for Reactor 3 (BF water + PAC), it was not necessary to change the sample inside the reactor, because of less SS accumulation. In the case of Reactors 2 and 3, particle size ranging from 1  $\mu\text{m}$  to less than 20  $\mu\text{m}$  shows the increasing tendency, but fluctuation shows that there is an increasing tendency even at 20

$\mu\text{m}$ . Inside Reactor 1, the particle size ranging from 1  $\mu\text{m}$  to less than 10  $\mu\text{m}$  sizes shows a sharp increasing tendency, while from 10–20  $\mu\text{m}$  sizes, it shows a decreasing tendency. But, during real observation, inside all reactors, the number of particles ranging from 1–10  $\mu\text{m}$  was higher. This size change may be due to the effect of continuous aeration, Brownian movement of particles and the impact of particles on wall of reactor and membrane. Figure 9 shows the SS accumulation rate inside the reactors. From this figure, it is clear that the SS accumulation inside Reactor 1 was the highest. But, inside Reactor 3 accumulation was minimum, because it was using biofilter treated river water. It is a very important effect of PAC that Reactors 1 and 2 were getting the same influent, but there is a big difference in retention rate with respect to the operation period. At the same time, the average accumulation rate of total bacteria cells followed the same pattern of SS retention that is shown in Figure 10, which gives a clear idea that with the successive retention of SS and gradual change of particle size inside the reactors, the number of total bacteria cells has also increased. It has already been demonstrated that colloidal particles carrying trace elements may be transferred to the particulate compartment (Stordal *et al.*, 1996). It is referred as “colloidal pumping”. However, the importance of suspended particulate process of pollutants might even extend beyond a simple carrier role. Organic matter, associated with suspended particulate and colloidal material, has been shown to be consistently different for both compartments (Rostad *et al.*, 1997), while a pumping of colloidal organic material or microorganism to the particulate phase is expected to homogenize the composition of the two phases. Such a discrepancy might be explained by the presence of bacteria within suspended particulate matter (Liss *et al.*, 1996; Paerl, 1975). The suction (TMP) and backwash pressure of the membrane modules are shown in Figures 11 and 12. During 162 days of operation, the membrane inside Reactor 1 fouled six times, whereas the membranes inside Reactors 2 and 3 fouled three times and twice, respectively. The fluctuation during backwash shows the time dependent variation of resistance to different adhesive materials onto the membrane surface. Here, filtrate was used to backwash the membranes. From the previous experience, it was found that membrane fouling in the PAC-MF process was due to cake and gel layer formation. When the microorganisms are attached on to the PAC and SS, the bulk volume of PAC is adsorbed on to the membrane and it caused the gel layer formation. These fine particles inside the reactor play a big role for membrane fouling with suspended PAC, attached or dispersed phase bacteria and retained SS.

### Conclusion

The development of a PAC-MF system for water treatment containing high concentration of PAC is one of the more-advanced technologies in the water industry. There is no clear evidence about the existence of microorganisms in the bulk phase. They might be in dispersed or attached phase (Khan *et al.*, 2001). By using PAC, the retention rate of SS can be decreased by a significant amount and PAC can also handle huge number of bacteria ( $5 \times 10^5$  cell/mL). But, when the system is without any PAC, it does not operate for long time even with less number of bacteria ( $2 \times 10^5$  cell/mL). The main reasons are the high accumulation rate of SS and gradual increment of finer particles. As a result of these complex phenomena, the membrane module inside Reactor 1 (raw water only) fouled the highest number of times, whereas Reactor 2 (Raw water + PAC) was getting same type of influent water, but fouled fewer times. The use of biofilter treated water as an influent can be appreciated for longer period of operation, which can handle the natural variation of SS as well as particles. It has also been proved that the PAC-MF system with backwash facilities was much better than that without backwash. Some effort should also be devoted to better understand the aggregate formation and dynamics of particles and bacteria, considering their role in geochemical cycling in natural waters and their pathways to the high dose PAC-MF system.

## References

- Clark, M.M. (1992). Ultrafiltration of lake water: Optimization of TOC removal and flux. In: *Influence and Removal of Organics in Drinking Water*, Chapter 23, Lewis Publishers, Boca Raton, Florida, USA.
- Clark, M.M. and Heneghan, K.S. (1991). Ultrafiltration of lake water for potable water production. *Desalination*, **80**, 243–249.
- Droppo, I.G. and Ongley (1992). The state of suspended sediment in the freshwater fluvial environment: a method of analysis. *Wat. Res.*, **26**, 65–72.
- Graham, M.R., Summers, R.S., Simpson, M.R. and Macleod, B.W. (2000). Modeling equilibrium adsorption of 2-methylisoborneol and geosmin in natural waters; *Wat. Res.*, **34**(8), 2291–2300.
- Khan, M.M.T., Ohgaki, S., Takizawa, S. and Katayama, H. (2001). Development of powdered activated carbon and microfilter membrane system for water treatment. *3rd International Summer Symposium of Japan Society of Civil Engineering (JSCE)*, 369–372.
- Lartigue, B.S., Mustin, S.D., Villemin, G., Mustin, C., Barres, O., Chamerois, M., Gerard, B., and Babut, M. (2001). Composition, structure and size distribution of suspended particulates from the Rhine river; *Wat. Res.*, **35**(3), 808–816.
- Lebeau, T., Lelievre, C., Buisson, H., Cleret, D., Van de Venter, L.W. and Cote, P. (1998). Immersed membrane filtration for the production of drinking water: combination with PAC for NOM and SOCs removal, *Desalination*, **117**, 219–231.
- Leppard, G.G., Flannigan, D.T., Mavrocordatos, D., Marvin, C.H., Bryant, D.W. and McCarry, B.E. (1998). Binding of polycyclic aromatic hydrocarbons by size classes of particulate in Hamilton harbor water. *Environ. Sci. Technol.*, **32**, 3633–3639.
- Lick, W. and Rapaka, V. (1996). A quantitative analysis of the dynamics of the sorption of hydrophobic organic chemicals to suspended sediments. *Environ. Toxicol. Chem.*, **15**, 1038–1048.
- Liss, S.N., Droppo, I.G., Flannigan, D.T. and Leppard, G.G. (1996). Floc architecture in wastewater and natural riverine systems. *Environ. Sci. Technol.*, **30**, 680–686.
- Paerl, H.W. (1975). Microbial attachment to particles in marine and freshwater ecosystems. *Microb. Ecol.*, **2**, 73–83.
- Perret, D., Newman, M.E., Negre, J.C., Chen, Y. and Buffle, J. (1994). Submicron particles in the Rhine river – I. Physico-chemical characterization. *Wat. Res.*, **28**, 91–106.
- Pontius, F.W. (1990). Complying with the drinking water quality regulations. *J. AWWA*, **82**, pp. 32–35.
- Rostad, C.E., Leenher, J.A. and Daniel, S.R. (1997). Organic carbon and nitrogen content associated with colloids and suspended particulates for the Mississippi and some of its tributaries. *Environ. Sci. Technol.*, **31**, 3218–3225.
- Seo, G.T., Ohgaki, S. and Suzuki, Y. (1997). Sorption characteristics of biological powdered activated carbon in BPAC-MF (biological powdered activated carbon-microfiltration) system for refractory organic removal. *Wat. Sci. Tech.*, **35**(7), 163–170.
- Stordal, M.C., Santschi, P.H. and Gill, G.A. (1996). Colloidal pumping: evidence for the coagulation process using natural colloids tagged with <sup>203</sup>Hg. *Environ. Sci. Technol.*, **30**, 3335–3340.
- Zabawa, C.F. (1978). Microstructure of agglomerated suspended sediments in northern Chesapeake bay estuary. *Science*, **202**, 49–51.