

B-30 Pilot Study on DOC and metal removal in powdered activated carbon-microfiltration (PAC-MF) water treatment process

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1. Introduction

Among the various membrane separation processes, membrane microfiltration (MF) filter is capable of satisfactorily removing particles, suspended solids, and bacteria from raw water under lower pressure. It, however, is ineffective in removal of colloids and dissolved organic matter due to its higher membrane-molecular-weight-cutoff (MMWC). High concentration of powdered activated carbon (PAC) coupled with MF membrane, *i.e.*, PAC-MF process, is considered to be a reliable and convenient water treatment process, which not only makes use of strong adsorption capacity of PAC to remove dissolved organic contaminants and helps biological oxidation of ammonia, manganese, and iron, successfully displaying the effective removal of particles and natural organic matter (NOM) in a single step, but also provides reduction of sludge volumes in treatment process. These advantages promote the research and application of PAC-MF process in a broad range of applications.

The objectives of this study were to verify the applicability of a noble PAC-MF process to the removal of DOC from raw water, which contains urban run-off and sewerage effluents; to evaluate the effect of the difference of PAC size and surface area on DOC removal rate; and to quantify the buffering effects of PAC-MF process against accidental contamination of raw water with metals and metalloids. To accomplish these objectives two pilot-scale reactors were operated in the Tamagawa Water Treatment Plant for 8 months.

2. Materials and Methods

2.1 Pilot experimental design and operational conditions.

Two pilot-scale PAC-MF reactors having the PAC adsorption zone (PAZ, 40L) and the micro-filtration zone (MFZ, 32L) were installed in the Tamagawa Water Treatment Plant, Tokyo. The raw water from Tamagawa River was pretreated through the biofilter and then sent to the PAC-MF reactors. The schematic flow diagram of the pilot experimental setup is shown in Figure 1. The mean PAC diameters were $36.5\mu\text{m}$ (Sirasagi C-1, Japan Environment Chemicals Co.) and $151\mu\text{m}$ (Shirasagi S-10) in Reactor-1 (R1) and Reactor-2 (R2), respectively. The PAC concentration in the PAZ was 20g/L and it was suspended by aeration (50 L/min). Tubular ceramic membranes (nominal pore size $0.1\mu\text{m}$, the total membrane surface area 0.174 m^2) were installed in the MFZ. Both membrane units were operated at the same flux ($42\text{ L/m}^2/\text{h}$ or 1 m/d). MFZ was aerated during backwash of membranes. The bulk PAC concentration in the MFZ varied from 2 g/L after backwashing to 0.2g/L at the end of each filtration cycle. The operational conditions of the pilot experiments in detail are shown in Table 1.

The real-time data of trans-membrane pressure (TMP), flux, and water temperature during the operations were transmitted to a personal computer through wireless PHS communication.

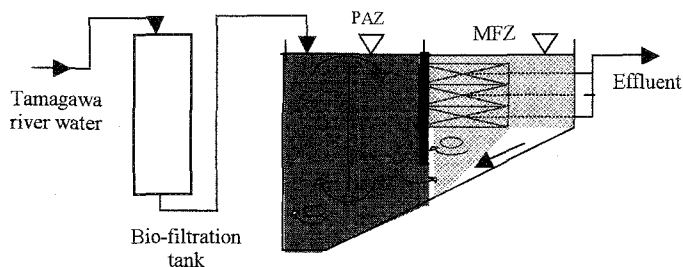


Figure 1. Schematic flow diagram of pilot PAC-MF system in Tamagawa Water Treatment Plant

2.2 Water quality analysis

Water samples were collected twice a week for the analyses of pH, DOC (TOC-5000A, Shimadzu, Japan), UV adsorption at 260nm (Spectrometer U-2010, Hitachi, Japan), and turbidity (Turbidimeter 2100N, Hach, USA).

Table 1. Pilot PAC-MF operational conditions.

Item				Reactor 1 (R1)		Reactor 2 (R2)	
Raw water source (Influent)				Biofilter pretreated raw river water			
PAC	Type			Shirosagi C-1		Shirosagi S-10	
	Material			Saw dust			
	Diameter (μm)	Mean		36.5		151.2	
		Median		24.9		139.7	
	Concentration			20g/L (based on PAZ volume)			
Membrane modules	Material			Ceramics			
	Surface area			0.058 (m ² /module)			
	Number of modules			3 modules in MFZ			
Filtration cycle				filtration (20 minutes) -- backwash (12 seconds)			
Filtration flux (m/d)				42 L/m ² /h or 1 m/d			
Hydro retention time (based on PAZ)				330 minutes			
Backwash	backwash flowrate			4m/d			
	backwash time			12 seconds			
Aeration	PAC Adsorption Zone			Throughout filtration (50L/min)			
	Micro-filtration Zone			During backwash (60L/min)			

2.3 Shock load test

After 8 months of operation six metal and metalloid ions were spiked into the PAZ of Reactor 2 to make the overall concentrations in the whole reactor ten times of the drinking water standards: namely, Cr (VI) -500 $\mu\text{g}/\text{L}$; Ni (II), As (III), Se (IV), Cd (II), Pb (II) - 100 $\mu\text{g}/\text{L}$. In the following four hours, the permeate samples were collected every 30 minutes, and their concentration were measured by ICP-AES.

3. Results and discussions

The operations of pilot PAC-MF processes were started on April 11, 2002. The two reactors showed different trends of TMP increase (Figure 2). At the beginning period, TMP of Reactor-1 with smaller PAC (C-1) remained in a lower level (6.0-8.0 kPa) for 20 days, then it went up to ca. 90 kPa after 82 days of operation (R1-Run1) due to PAC cake layer formation. Then the fouled MF membrane modules were taken out to remove the PAC cake fouling attached on membrane surface by scraping with sponge and water. The membranes were then re-installed in R1 for second period of operation (R1-Run 2), which lasted for 45 days (July 3-August 18) until the MF membranes were fouled once again. The fouled membranes were physically cleaned in the aforementioned method, and used PAC in the bulk solution in R1 was chemically cleaned by 2% citric acid to extract metal ions adsorbed in the earlier operation. The third operation (R1-Run 3) was commenced on Aug 20 and it lasted 33 days (until September 22). However, the TMP change of the reactor with larger PAC (R2, S-10) demonstrated quite a different tendency. It reached a relatively stable trans-membrane pressure (TMP) of 40-50 kPa after 40 days of operation, thereafter remained in a quasi-steady value for nearly 5 months. In late October TMP in R2 increased due to low water temperature, although the PAC cake resistance was relatively stable. The operation of R2 was finally stopped in December 26, 2002 at a TMP of 84 kPa after 260 days of operation.

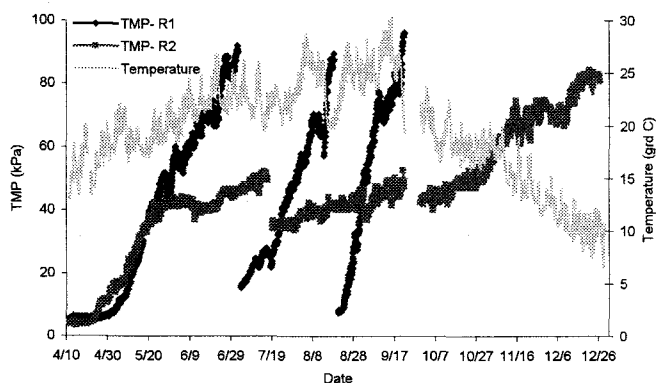


Figure 2. TMP and water temperature change during pilot PAC-MF operations

Figure 3 (left) shows DOC values of the influent, the bulk solution in reactor, and the effluent for both R1 and R2 reactors. Their DOC removal rates during the whole operation are also shown in Figure 3 (left). Figure 3 (right) displays the average removal rates of DOC and the average turbidity of influent and effluent for each reactor. Both reactors possessed relatively high organic removal rates (85-90%) at the beginning period of operation, while decreased gradually with operational time. It is also noted that a significant DOC removal rate, *ca.* 10-15%, was achieved in MFZ though the PAC concentration there was much lower than in PAZ. This may be explained by the following mechanisms: (1) the water flow regime in the PAC cake layer on the membranes was plug-flow, which demonstrates a higher removal rate than completely mixed reactors; (2) the average PAC particle sizes was lower in MFZ, wherein larger PAC particles were separated by sedimentation; and (3) the PAC cake layer provided a higher opportunity for the water to contact PAC surface.

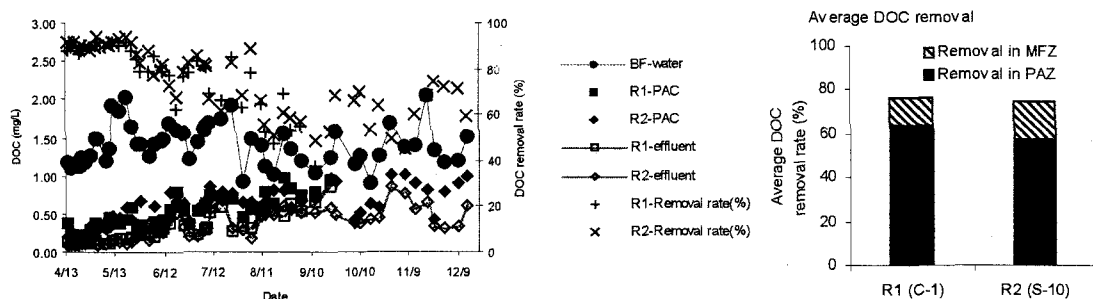


Figure 3. DOC and its removal with time during the pilot PAC-MF operations (left) and the average DOC removal rates (right).

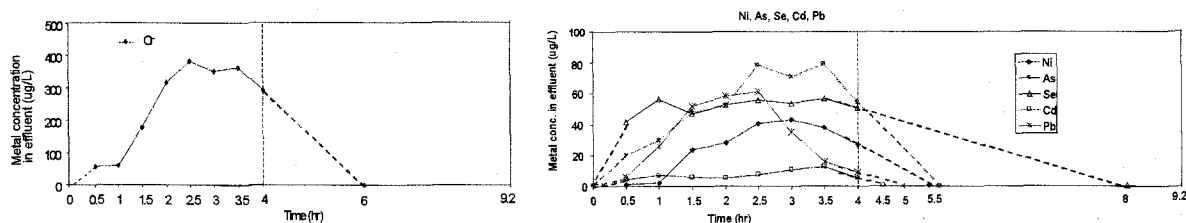


Figure 4. The concentration change of heavy metal ions in R2-effluent during shock load test

Figure 4 demonstrates the concentration of heavy metal ions in effluent of R2 after the spike of these elements. This shock-load test was carried out on December 18, 2003, nearly at end of the operation. Due to operational regulations samples were taken only up to 4 hours after the spike. The metal concentrations in the effluent were linearly extrapolated after 4 hours, and by comparing the difference between the doses of metals spiked into reactor and their weights in effluent (mass balance calculation) so that the overall removal rates were estimated, which are: Cd(96%) > Ni(87%) > Pb(86%) > Cr(75%) > As(75%) > Se(69%). As the concentrations of these metals were set at ten times higher than the drinking water standard, the goal of removal rates was 90%. Only one metal species, cadmium, reached over 90% removal, whereas two more metals, lead and nickel, had nearly removal rates slightly less than 90%. The removal rates were lower for three other metals, but it was demonstrated that after 8 months of operation PAC-MF process has a certain buffering capacity in case of accidental contamination of raw water by these metals.

4. Conclusions

The pilot PAC-MF process demonstrated very high organic removal rates (85-90%) for two months in the beginning of operation, then gradually decreased to *ca.* 60%. The overall removal rates were 76% for small PAC (C-1) and 74% for the large PAC (S-10). The differences in PAC sizes and BET surface area did not cause any difference in DOC removal rates. In spite of low PAC concentration and shorter HRT 10-20% of organic removal took place in MFZ, which demonstrates the effectiveness of separating the adsorption zone (PAZ) and the membrane separation zone (MFZ). The metal shock-load revealed that metals were adsorbed on PAC in the order of Cd > Ni > Pb > Cr > As > Se, which demonstrated buffering capacity of PAC-MF process in case of accidental metal pollution.