



## Removal of high-concentration geosmin using a GAC filter and a cross-flow-type membrane filtration process combined with PAC

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### ABSTRACT

The purpose of this study is to remove high-concentration geosmin (GSM) by applying a granular-activated carbon (GAC) filter and a membrane filtration (MF) process injected with powdered-activated carbon (PAC). In this study, laboratory-scale experiments were conducted to (1) develop isotherm equations for GSM adsorption using PAC and GAC, (2) assess GSM adsorption competition by dissolved organic carbon (DOC), and (3) derive design factors for a pilot plant. In addition, the flux of the cross-flow-type pressurized MF was assessed at various PAC concentrations. Based on the results, a pilot plant was designed and operated from 14 July to 16 August 2013. The pilot plant operation achieved not only a stable treated water quality with an average GSM concentration of 10 ng/L or less (removal efficiency: 99.5%), but also a treated water quality with an average DOC concentration of 0.5 mg/L or less (removal efficiency: 91.7%) at the same time. From these findings, it was concluded that it may be possible (a) to meet the monitoring criteria for drinking water quality when high-concentration GSM is exposed, (b) to reduce the total amount of PAC inputs, and (c) to redress possible public complaints about compounds with unpleasant taste and odor.

*Keywords:* High-concentration geosmin; Cross-flow-type membrane filter; Powdered activated carbon; Granular activated carbon

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### 1. Introduction

Climate change has caused various abnormal events including reduced precipitations and rising temperatures. This especially affects water resources and creates problems in drinking water supplies. Among such problems, abnormal taste and odor substances (T&Os) in drinking water are one of the main causes of repelling consumers from tap water [1]. It

has been reported that the occurrence of T&Os is attributable to disinfectants added in the process of water purification and to chemicals contained in sewer and wastewater. In particular, earthy and musty smells produced by geosmin (GSM) and 2-methylisoborneol (2-MIB) in the metabolic process of *cyanobacteria* (blue-green algae) and *actinomycetes* are suggested as a major cause of T&Os [2]. In South Korea, GSM has been known as a key algal metabolite that generates earthy and musty smells. In summer 2012, Korean

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drinking water supplies were exposed to a 4,400 ng/L of GSM, which far exceeds the monitoring criteria for drinking water quality (20 ng/L). This caused water purification problems and public complaints about drinking water. GSM is a tertiary alcohol which imparts earthy–musty T&Os in drinking water and can be detected by consumers at as low concentrations as 5–10 ng/L. Such algal metabolites are known to be difficult to treat in the conventional way of water treatment, especially when they are in a dissolved form [3–5].

The processes of coagulation/sedimentation/filtration included in the conventional treatment facilities are very effective in removing algal cells, but not effective in eliminating the GSM produced from those cells [6,7]. To remove the dissolved metabolites efficiently, it has generally been suggested to introduce additional oxidation and adsorption processes. Ferguson et al. [8], Glaze et al. [9], and Bruce et al. [10] eliminated GSM using ozone, hydrogen peroxide, and UV. The addition of chemicals, however, is expensive and can generate disinfection by products (DBPs), which are unacceptable in human health and regulatory terms.

Adsorption by granular-activated carbon (GAC) or powdered-activated carbon (PAC) can be applied as one of the best available techniques for removing T&Os and organic substances contained in drinking water. Many studies report findings of GSM adsorption using PAC and GAC [11–16]. However, PAC and GAC adsorption can be inhibited by constituents in polluted raw water, especially dissolved organic carbon (DOC) [17–22]. DOC consists of a mixture of organic compounds with various chemical characteristics. It has a wide range of molecular weights and can be adsorbed by activated carbon. In addition, DOC can be adsorbed to larger pores, thereby blocking the adsorption of GSM which prefers attaching to thermodynamically smaller surfaces [23–25].

The removal of DOC in advance using activated carbon can improve GSM adsorption efficiencies [19,26]. In recently published papers, the membrane filtration (MF) process and GAC filters are applied to remove DOC and T&Os. Still, there are not sufficient design factors for GSM treatment. Moreover, the existing literature focuses only on the removal of relatively low-concentration GSM (Table 1).

In this view, this study was conducted to determine how to satisfy the monitoring criteria for GSM concentration in drinking water which has rapidly increased in Korea since 2012. This study was also aimed at an efficient removal of high-concentration GSM using a GAC filter and a cross-flow-type MF process fed with PAC.

Table 1  
Initial GSM concentrations in the existing literature

Literature	Water source	GSM ng/L
Ho et al. [27]	Morgan WTP, South Australia	200
Kim et al. [28]	Suji WTP, Korea	70
Drikas et al. [1]	Mt P. WTP, South Australia	200
Lin et al. [29]	Laboratory reagent water	200
Cook and Newcombe [30]	Laboratory reagent water	300
Graham et al. [31]	Laboratory reagent water	170
Yuan et al. [32]	Laboratory reagent water	500
Zoschke et al. [33]	Laboratory reagent water	100
Chen et al. [34]	Laboratory reagent water	200

## 2. Materials and methods

### 2.1. Materials and reagents

Norit PAC200 coal-based PAC and Norit GAC1240 coal-based GAC (Cabot, USA) were washed with purified water 10 times to remove PAC and GAC fines prior to use. Solid GSM (98%) used in this study was purchased from Wako Pure Chemicals, Ltd. (Osaka, Japan). Sterile GSM solution (20 mg/L) was prepared by dissolving GSM in purified water treated with a Milli-Q system (Samwoo S&T Ltd., Korea). Potassium hydrogen phthalate ( $C_6H_4(COOK)(COOH)$ , Nacalai Tesque, Japan) was used as a standard reference for DOC. The glassware was washed three times with purified water, and then, sterilized at 121 °C for 30 min before use.

### 2.2. Laboratory experiments

Laboratory-scale experiments conducted in this study consist of three parts: (1) determine GSM removal efficiencies with different reaction times and PAC inputs to set the contactor's retention time, (2) develop isotherm equations for GSM adsorption using PAC and GAC, and (3) assess DOC interference with GSM adsorption. The experimental conditions are described in Table 2.

The Freundlich model was used to assess GSM adsorption and DOC interference with GSM adsorption. As one of the most popular adsorption models for a single solute system, the Freundlich model is an empirical equation based on the distribution of solute between the solid phase and aqueous phase at equilibrium [35]. The basic Freundlich equation is:

$$x/m = K_f C_e^{1/n} \quad (1)$$

Table 2  
Laboratory-scale experiments for removing high concentrations of GSM

Lab. test	Experimental conditions	
(i) PAC contactor time	Reaction time, min	
	7.5	30 45 60
(ii) GSM adsorption isotherms	PAC and GAC dosage, mg	
	1.0 1.5	2.0 2.5 4.0
(iii) DOC adsorption inhibition	Add DOC conc., mg/L	
	0.5	1.5 2.0 2.5

where  $x$  is the amount of GSM adsorbed;  $m$  is the weight of carbon;  $x/m$  is the concentration of GSM adsorbed;  $C_e$  is the equilibrium concentration of GSM in solution; and  $K_f$  and  $n$  are the empirical constants. Eq. (1) can be rearranged into a linear form:

$$\log(x/m) = \log K_f + (1/n) \log C_e \quad (2)$$

For an X-Y plot of Eq. (2), where  $y = \log(x/m)$  and  $x = \log C_e$ , the slope is  $1/n$  and intercept is  $\log K_f$ .

### 2.3. Permeate flux of the membrane with PAC injection

In order to maximize GSM adsorption rates without any additional PAC settlement tank installed, PAC contact water was flowed into a MF and circulated until it reached the adsorption breakpoint (pre-test for pilot plant operation). The MF employed in this study is made by Aquasource (L-PSF / PS300, France), a cross-flow-type of pressure-resistant hollow fiber membrane. The nominal pore size is 0.02, and the effective filtration area is 93 m<sup>2</sup>/module. The permeate flux ranges between 1.4 and 3.6 m<sup>3</sup>/m<sup>2</sup>d (60–150 LMH). Two modules were installed in Yangse water treatment plant (WTP) in South Korea to measure permeate fluxes with different PAC inputs. The measurement conditions of MF flux with varying PAC inputs are shown in Table 3.

### 2.4. Pilot-scale experiments

A pilot-scale reactor was built based on the design factors derived from the laboratory-scale experiments

and the assessment of membrane permeate fluxes with various PAC inputs. First, a PAC contactor was installed to circulate PAC inside the cross-flow-type of membrane to remove high-concentration GSM. The treated water filtered from the membrane process was designed to finally discharge through a GAG filter. A constant amount of PAC (min. 78.6 mg/min, max. 209.3 mg/min) was injected into the PAC contactor. Two sets of GAC filters were built: One of them was for the operation and the other was as a spare. The reason that the GAC filter was placed after the PAC-MF process was because GSM adsorption is inhibited by DOC. The GAC filter process was intended to improve GSM adsorption efficiencies after a certain amount of DOC was removed in the PAC-MF process. Fig. 1 shows the pilot-scale plant for removing high-concentration GSM.

### 2.5. Analysis of GSM and DOC

The solid-phase micro-extraction (SPME) was applied to adsorb volatile substances to fibers, and the GC/MS was used to measure GSM concentrations (GC: Varian CP-3800, MS: Varian Saturn 2200, SPME fiber: polydimethylsiloxane/divinylbenzene). The limit of quantification was 1 ng/L. After samples were filtered through a 0.45 membrane, the multi N/C 3000 (Analytik Jena AG, Germany) was used to make the non-purgeable organic carbon (NPOC) measurement of DOC concentrations. Many studies report that the GSM concentration has a narrow range of fluctuations (5–10%) at room temperature, indicating that there is little effect from temperature upon GSM concentrations [36–39]. Before being analyzed, however, the

Table 3  
The experimental conditions of membrane flux with PAC dosage

Raw water characteristics at Yangse WTP (2013. 5~6)	Temp., °C 17.2 ± 3.3	pH 7.1 ± 0.4	DOC, mg/L 3.5 ± 0.4	Turbidity, NTU 5.6 ± 2.5
Membrane evaluation	Permeate flux (LMH, L/m <sup>2</sup> hr)			
PAC dosage, mg/L	0, 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5, 4.0, and 5.0			

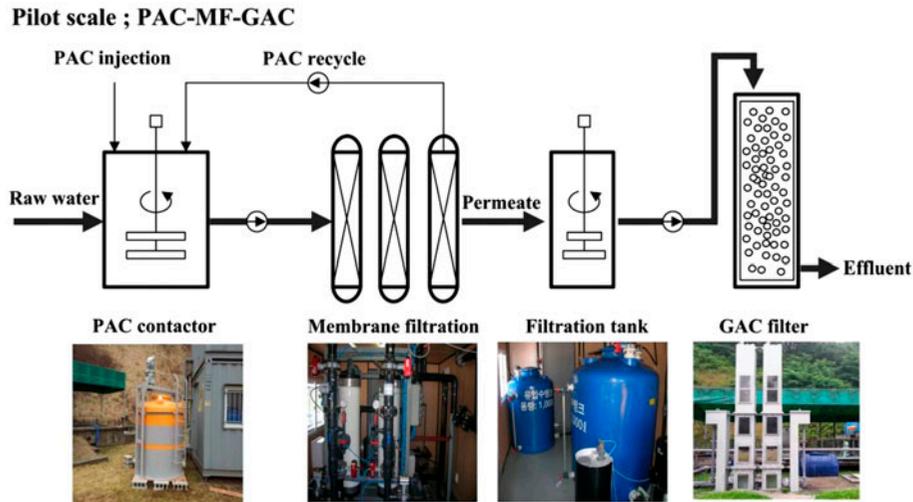


Fig. 1. PAC-MF-GAC process of high-concentration GSM removal.

samples were covered with a cap immediately after sampling in order to prevent the volatilization of GSM.

### 3. Results and discussion

#### 3.1. Removal of high-concentration GSM using PAC and GAC on a laboratory scale

Fig. 2 shows the result of removing GSM with different PAC contact times on a laboratory scale. The result indicates that GSM concentrations declined along with longer contact times. In addition, higher PAC inputs at various contact times led to a linear decline in GSM concentration under all conditions. At contact times of 45–60 min (PAC input: 15 mg/L), GSM removal efficiencies reached 61–66%, respectively, showing no significant difference between them. The PAC contact time to remove high-concentration GSM, therefore, was set as 45 min. In the existing literature, removal efficiencies of more than 95% are achieved with the PAC contact time and input set as 30 min and 30 mg/L at influent GSM

concentrations of 37–100 ng/L [28,30]. To eliminate high-concentration GSM using PAC more efficiently, the contact time may need to be longer than the times suggested by the existing literature.

Fig. 3 shows the results of GSM adsorption isotherm equations for PAC (Fig. 3(a)) and GAC (Fig. 3(b)) when DOC was not added. The isotherm equations for PAC injection were  $16.00C_e^{0.69}$ ,  $15.78C_e^{0.64}$ , and  $24.21C_e^{0.72}$ , respectively, while those for GAC injection were each  $21.38C_e^{0.66}$ ,  $31.92C_e^{0.58}$ , and  $23.44C_e^{0.64}$ . The isotherm equations for PAC and GAC averaged for the three replicate experiments were  $18.66C_e^{0.68}$  and  $25.58C_e^{0.63}$ , respectively. All *P* values satisfied the criteria of 0.05 or less. Thus, the results were found significant. The experiments indicated that GAC has a higher adsorption capacity than PAC. This may be because GAC used in this study is higher than PAC in iodine number which determines adsorption capacity (PAC: 900 mg/g, GAC: 1,000 mg/g). Values of  $K_f$  and  $1/n$  in the existing literature are shown in Table 4 as follows. In this study,  $K_f$  values were lower than those suggested by the existing literature. Based on these results, it was

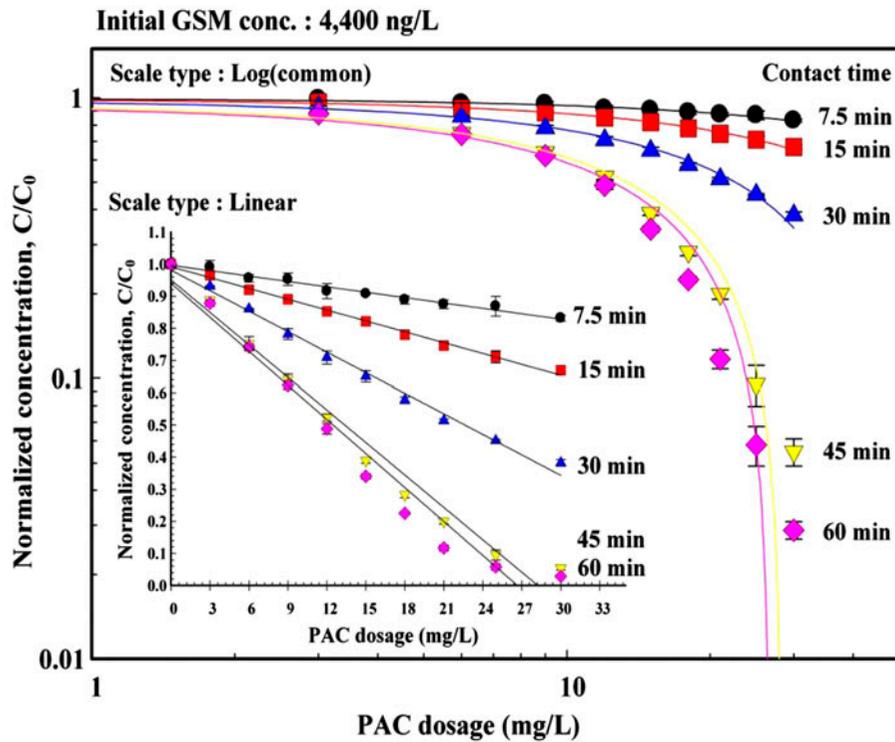


Fig. 2. Result of removing GSM with different PAC contact times on a laboratory scale.

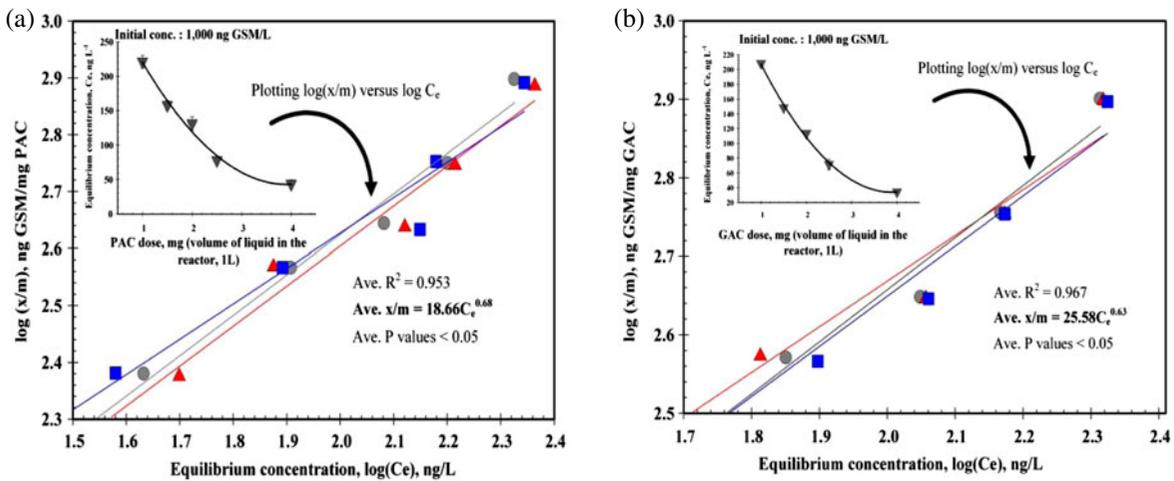


Fig. 3. GSM adsorption isotherm equations for PAC and GAC: when DOC was not added, (a) Adsorption isotherms of GSM (PAC) and (b) Adsorption isotherms of GSM (GAC).

concluded that adsorption rates dropped with higher concentrations of GSM.

The adsorption capacity for T&Os was assessed for the application of PAC and GAC in a real treatment plant, and the result is shown in Fig. 4. In the adsorption competition with DOC, higher  $K_f$  reduction rates were observed with GAC (25.58 with no input, 0.03

with an input of 2.5 mg/L) than PAC (18.66 with no input, 0.07 with an input of 2.5 mg/L). The reason may be because DOC is adsorbed to larger pores and interferes with the adsorption of GSM, which prefers attaching to thermodynamically smaller surfaces [12,23–26]. In a study by Choi on adsorption competition between DOC and T&Os, the input of DOC

Table 4  
Comparison of adsorption capacity between this study and the existing literature

Items	Literature				This study	
	[33]	[31]	[13,14]	[40]	PAC	GAC
Initial GSM conc., ng/L	100	170	100	50	1,000	1,000
$K_f$ , (mg/g)/(mg/L) <sup>n</sup>	137	45.6	280	26.6	18.6	25.6
$n$	0.15	0.28	2.04	2.04	1.47	1.59

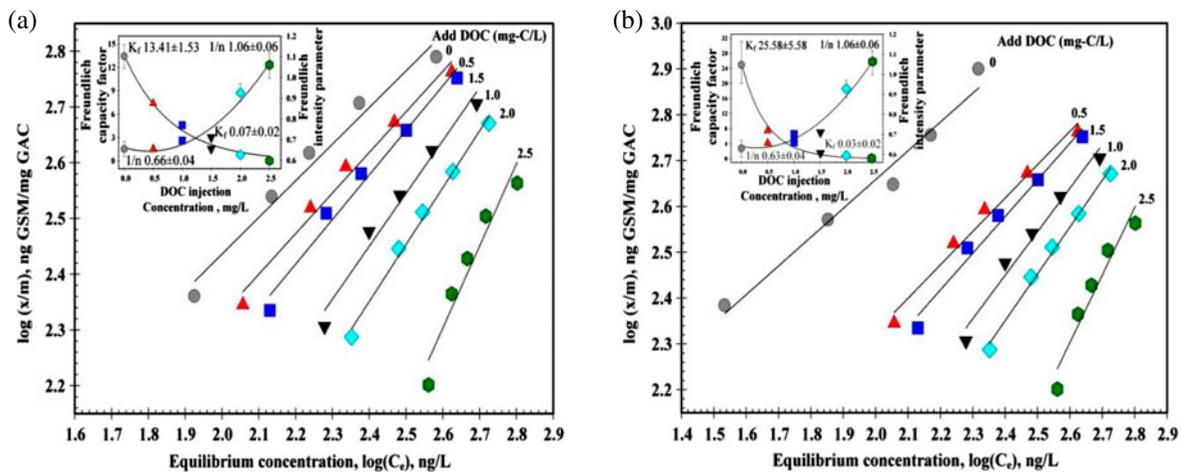


Fig. 4. GSM adsorption isotherm equations for PAC and GAC: when DOC was added, (a) Adsorption isotherms of GSM (PAC) and (b) Adsorption isotherms of GSM (GAC).

resulted in an adsorption capacity 100 times lower than when DOC was not added [40]. Many studies also suggest that DOC affects the adsorption of T&Os [10–12,16,30]. Therefore, it is desirable to consider DOC competition in adsorption when designing a pilot plant on site for high-concentration GSM removal.

### 3.2. Assessment of MF fluxes with different PAC inputs

The MF fluxes with various PAC injections showed that the flux dropped in a quadratic form ( $\text{Flux} = -2.0530X^2 - 5.3545X + 104.85$ ) when PAC input increased (Fig. 5). With no PAC injection, the flux averaged 106.75 LMH, whereas with a PAC input of 5 mg/L, the flux fell to 29.33 LMH on average. In particular, the flux sharply dropped when 3.5 mg/L of PAC was injected. According to Kim and Bae, who investigates PAC effect on the MF process, LMH decreases by about 50% when the PAC injection is 50 mg/L [41]. Tomaszewska and Mozia reports that LMH dropped by 60% with a PAC input of 10 mg/L [42].

### 3.3. Assessment of high-concentration GSM removal using the PAC-MF-GAC process in a pilot-scale plant

Table 5 shows design values for pilot plant establishment based on the results of the laboratory-scale and PAC/MF experiments. The pilot plant was equipped with a PAC-cross-flow-type MF-GAC filter.

Fig. 6 shows influent GSM concentrations and PAC injections in the period of pilot plant operation (July–August 2013). During the operation, GSM flew in at an average of 1,712 ng/L, and the highest influent concentration was 4,256 ng/L, which indicates that GSM concentrations during the pilot operation were as high as in 2012. The PAC input concentrations were set based on the daily analysis of GSM concentrations in the settlement tank water. The amounts of PAC inputs ranged from 78.6 to 209.3 mg/min with different GSM concentrations. When influent GSM concentrations were below 1,000 ng/L, which were designed for the pilot operation, no additional PAC was injected.

The average GSM removal efficiency with PAC inputs and the MF process during the pilot operation

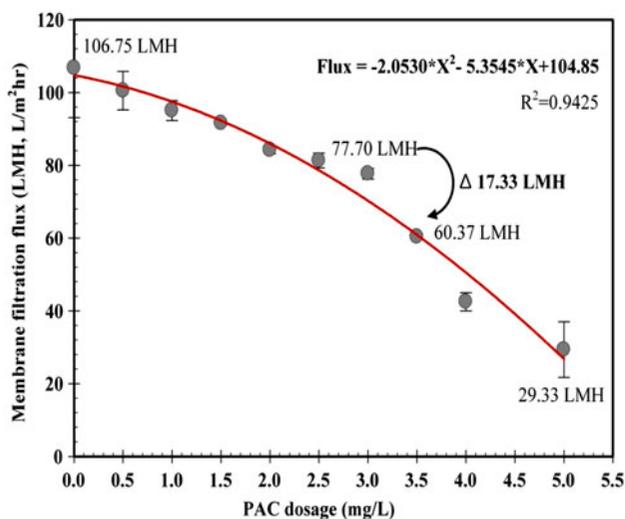


Fig. 5. MF flux variations with different PAC injections.

was found to be 51.7%. Before the influent flowed into the GAC filter, the average concentration of GSM was 826.6 ng/L. After it was discharged from the GAC filter, the average GSM concentration was 8.67 ng/L (removal efficiency: 99.5%). This indicates that it can satisfy the monitoring criteria for drinking water quality (20 ng/L in Korea) (Fig. 7(a)). As the findings indicated, GSM removal efficiencies in the PAC-MF process were low (51.7% on average). This may be

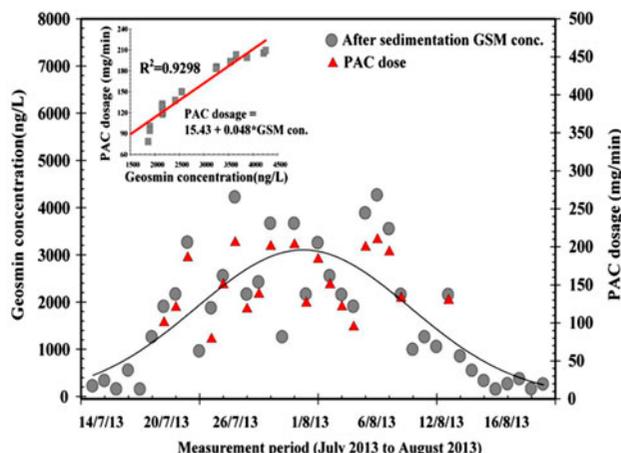


Fig. 6. Influent GSM concentrations and PAC injections during the pilot operation of PAC-MF-GAC process.

because of competition between GSM and DOC for adsorption. To remove high-concentration GSM more efficiently, therefore, it may be necessary to remove DOC in advance before adsorbing GSM. The existing studies suggest that T&Os can be effectively removed with the application of PAC, GAC, membranes or ozone [12–14,43–45]. However, these studies are mostly based on experiments with low influent GSM concentrations. It might be hard to remove high-concentration GSM using either PAC or GAC alone.

Table 5

Design of a pilot plant based on laboratory-scale, PAC and GAC, and MF experiments

Pilot plant design		Value	Unit
PAC	PAC contact time	45	min
	PAC dosage <sup>a</sup>	< 3.0	mg/L
	$K_f$	0.065	(ng/mg)(L/ng) <sup>1/n</sup>
	$n$	0.87	–
M.F.	Flux <sup>b</sup>	60–107	LMH
GAC filter	Influent GSM design conc.	1,000	ng/L
	Flow rate	6.94	L/min
	EBCT, Empty bed contact time	15	min
	$V_f$ , Linear approach velocity	135	m/d
	Carbon density	450	g/L
	Carbon require <sup>c</sup>	46,875	g
	$K_f$	1.48	(ng/mg)(L/ng) <sup>1/n</sup>
	$n$	1.46	–
	CUR, Carbon usage rate <sup>d</sup>	0.0045	g GAC/L
	Volume of water treated <sup>e</sup>	10,405,887	L

<sup>a</sup>PAC injections are set after settlement tank water is measured.

<sup>b</sup>PAC concentrations inside MF are maintained at 3.0 mg/L or less.

<sup>c</sup>Carbon require: EBCT × Q × ρ<sub>GAC</sub>, 15 min × 6.94 L/min × 450 g/L.

<sup>d</sup>CUR:  $(C_0 - C_e) / K_f C_0^{1/n} = (1,000 - 5) \text{ ng/L} / (1.48 \times 1,000^{0.706})$ .

<sup>e</sup>Volume of water treated: Mass of GAC for given EBCT/GAC usage rate = 46,875 g / 0.0045 g GAC/L.

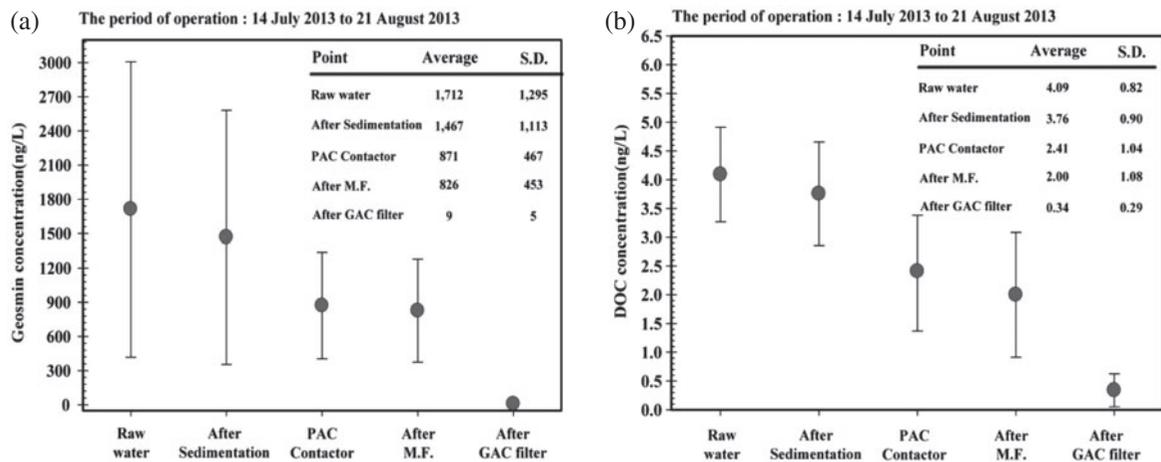


Fig. 7. GSM and DOC concentrations in each unit process during the pilot operation: (a) Removal of GSM and (b) Removal of DOC.

Moreover, the factors that were designed for removing low-concentration GSM in the existing literature may not be appropriate for removing high-concentration GSM.

During the pilot operation, the influent DOC concentration averaged 4.09 mg/L (Fig. 7(b)). In the GAC-filtered water, the average DOC concentration was 0.34 mg/L, and the removal efficiency was 91.7%. Omer et al. [46] report a DOC treatment efficiency of about 60% in PAC and MBR processes. Ho and Newcombe [16] also show a DOC removal efficiency of around 56% in experiments of NOM removal using PAC. In this study, the DOC removal efficiency reached more than 90% when both PAC and GAC processes were applied at the same time to remove high-concentration GSM. High DOC removal efficiencies may be a solution to the problem of disinfection byproducts.

#### 4. Conclusions

In this study, a pilot plant operation followed by laboratory-scale tests was performed to determine on-site applicable PAC inputs, necessary GAC amounts, and availability. In addition, adsorption capacities were derived from adsorptive competition with DOC. The result of the pilot plant operation indicated that it may be possible to achieve a stable treated water quality with an average GSM concentration of 10 ng/L or less (removal efficiency: 99.5%) and an average DOC concentration of 0.5 mg/L or less (removal efficiency: 91.7%) at the same time. The cross-flow-type MF process may also allow the application of PAC injection alone without having to use additional settlement faci-

ties. Moreover, PAC consumption can be reduced through the cross-flow filtration and calculation of an optimal PAC input. Based on the findings of this study, it was concluded that it may be possible to (a) meet the monitoring criteria for drinking water quality when high-concentration GSM is exposed, (b) reduce the total amount of PAC inputs, and (c) redress possible public complaints about T&Os. The findings of this study may also provide a practical solution for an efficient treatment of GSM, a rapidly increasing T&O in recent years.

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