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Removal of geosmin with inter-chlorination using powdered activated carbon in full-scale WTP

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ABSTRACT

The occurrence of algal and Taste and odor compounds was investigated in Han River, Korea. Large numbers of algae flourish in a water body. The geosmin concentration increases to levels of 378 ng L^{-1}. It was very difficult to control the geosmin through powdered activated carbon (PAC) treatment only. In this study, to remove the high concentration of geosmin, inter-chlorination treatment was applied between sedimentation basin and filtration. The removal efficiency of geosmin was evaluated through combined treatment by PAC and chlorination. The exclusive inter-chlorination treatment was more effective than combined treatment by pre-chlorination and inter-chlorination. It was proved to be removed above two times at low geosmin levels and 1.2 times at medium geosmin levels. Particularly, in the season of cyanobacterial blooms, the removal efficiency of geosmin was increased to 94%. Thus, the combined treatment by PAC and exclusive inter-chlorination has a synergy effect to remove geosmin.

Keywords: Inter-chlorination; Algae; T&O compounds; geosmin; PAC

1. Introduction

Taste and odor (T&O) have long been associated with the suitability and safety of our drinking water. Most T&O events in water lakes worldwide are caused by secondary metabolites of cyanobacteria, mainly geosmin and 2-MIB. These can be detected by consumers as musty-earthy odor at levels as low as 10 ng L^{-1} [1]. To preserve the esthetic quality of drinking water, the concentration of the T&O compounds has to be reduced below the threshold odor concentration by an effective elimination process. The treatment with powdered activated carbon (PAC) has proved to be efficient for the removal of geosmin and 2-MIB as it was relatively inexpensive and can be applied only when required [2].

Currently PAC is the most commonly practiced technology for 2-MIB/geosmin removal. This was mainly because the T&O episodes are seasonal and PAC offers the treatment plants with the flexibility to turn on/off dosages and also to adjust dosages depending upon the severity of the event [3]. The main steps of Amsa water treatment plant (WTP) in Korea, the subject of this study, are intake station, gagging well, coagulation/flocculation, sedimentation, sand

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filtration, and disinfection. In case of the seasonal occurrence of T&O compounds in the raw water, a PAC suspension was added at the gaging well. Recently, in the raw water of Han River in Korea, large numbers of algae flourish in a water body. The geosmin concentration increases to levels of 378 ng L^{-1}. This level was 38 times of threshold concentration. Some study shows that to reduce 40 ng L^{-1} of geosmin to 10 ng L^{-1} at four raw waters given a contact time of 50 min, minimum PAC dose was required as 21–29 mg L^{-1} [4]. Also, after 20 mg L^{-1} of PAC was added to the pilot-scale horizontally baffled PAC contactor at 1, 10 min contact time, the geosmin removal rate reached 40 and 76% [5].

When geosmin concentration flows in above 80 ng L^{-1}, it was very difficult to control the geosmin through only PAC treatment. And, in short time, establishing advanced treatment process such as granular activated carbon or O_3 was not easy.

Chlorination was one of many methods that can be used to disinfect water. Pre-chlorination practice has been conducted to remove ammonia and organic matters in raw water. The pre-chlorination treatment can increase geosmin levels through destroying the cells of cyanobacteria when cyanobacteria outbreaks. In the present study, to effectively reduce geosmin, the inter-chlorination treatment was applied between the sedimentation basin and sand filtration.

The goal of this study was to evaluate the removal efficiency of geosmin by combined treatment with inter-chlorination and PAC. Firstly, after estimating characteristics of species of algae during the study period, a suitable process was proposed. Secondly, after correlation of cyanobacteria and geosmin was analyzed, the ratio of particle-bound geosmin and dissolved geosmin was estimated. Finally, to treat the low and medium geosmin levels, the removal efficiency of geosmin was evaluated with ratio of pre-chlorination and inter-chlorination. The effective treatment reducing geosmin was suggested when high concentration of geosmin flows in.

2. Materials and methods

2.1. Study site and WTP process

The Han River is a major river in South Korea with a length of 514 km. As illustrated in Fig. 1, it is formed by the confluence of the south Han River and north Han River. Two major branches of the river come together at Yangsu-ri, Gyeonggi-do province, and that point is referred to as the Han River. It then passes through Seoul. Amsa WTP, study site, is located in the Han River as shown in Fig. 1.

Raw water sample was taken from the Han River, Seoul, Korea. The raw water quality over the study period from January to December 2012 consisted of turbidity ranging from 3 to 229 nephelometric turbidity units, algae from 430 to 22,330 cell counts, and geosmin from 1.0 to 378 ng L^{-1}. As illustrated in Fig. 2, Amsa WTP was operated as conventional treatment solution were found to be/flocculation, sedimentation, sand filtration, and disinfection.

The pre-chlorination and PAC treatment was applied at intake station and gaging well, respectively. The inter-chlorination treatment was applied between sedimentation basin and sand filtration.

2.2. Materials

Geosmin was obtained from SUPELCO, a division of Sigma-Aldrich Corporation. The 2-MIB and Geosmin MIX solution were found to be >99.9% pure by GC/MSD analysis.

PAC was supplied by Union Carbon Co., Ltd. This activated carbon was known to be mainly microporous, with very little meso or macroporosity.

2.3. Methods

2.3.1. Counting of algae cells

After algae were sampled from Amsa intake station, they were collected in an approved 500 ml polyethylene container. A preservative (Lugol's solution) is added to maintain algal cell structure for reducing microbial decomposition. All samples were kept cool and out of direct sunlight.

Cell counts of algae were determined by a Sedgwick-Rafter Cell S50 (SPI Supplies) using an inverted Axiovert 200 microscope (Zeiss).

2.3.2. Analysis of T&O compounds

A GC/MSD (5975C, Agilent Technologies, USA) was employed for the analysis of geosmin. The injection port was operated in splitless mode with temperature controlled at 250℃ and pressure at 7.6 psi. The oven temperature was held at 50℃ for 5.0 min, followed by an increase at a rate of 15℃/min to 65℃ and held for 5 min. Then the oven temperature was further increased to 220℃ at a rate of 15℃/min, and held for 1 min. The calibration curves for geosmin were established between 0 and 500 ng L^{-1} for PDMS fiber, with high linearity and high regression coefficients ($R^2 > 0.999$).

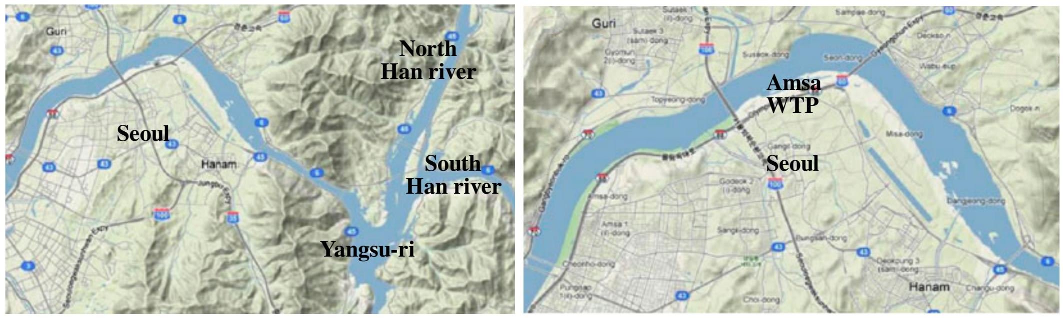

Fig. 1. Map of Han River located in Seoul, Korea.

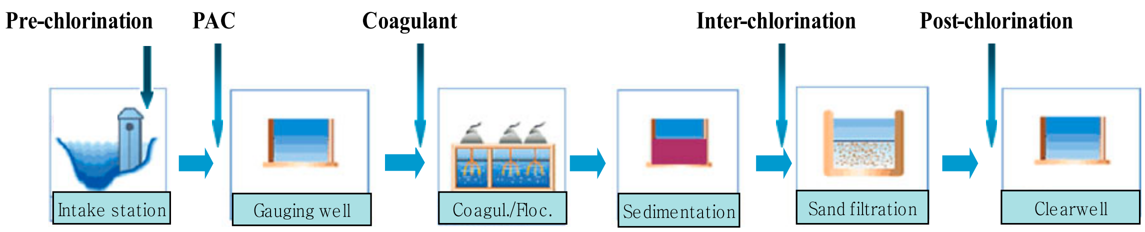

Fig. 2. Conventional treatment process of Amsa WTP.

3. Results and discussion

3.1. Effective WTP operation with variation of dominant algae

Dominant algae species were different with varying seasons. Fig. 3(a) shows algae cell counts over the study period from January to December 2012. Fig. 3(b) indicates the variation of algae and temperature during algae bloom season of 2012.

As shown in Fig. 3(a), total algae were occupied as diatom all year-round except for August. Diatom consists of *Cyclotella*, *Melosira*, and *Synedra*. Diatom flourished from 3,320 cell counts to 21,790 cell counts.

Diatom dominates phytoplankton communities from the start of spring season to winter season except for summer season. Diatom typically exhibit in freshwater environment. When conditions in the upper mixed layer which is rich in nutrients and light are favorable, their competitive edge allows them to quickly dominate phytoplankton communities [6].

However, the diatom usually causes filter-clogging in the water treatment process. The filter duration time can be reduced by clogging of diatom. So, in the season of diatom blooms, it is necessary to apply pre-chlorination to intake station. The pre-chlorination treatment prevents the filter-clogging from destroying the diatom cells.

As illustrated in Fig. 3(b), during the period of July–August, the dominant of algae was changed to cyanobacteria. As soon as the temperature increases

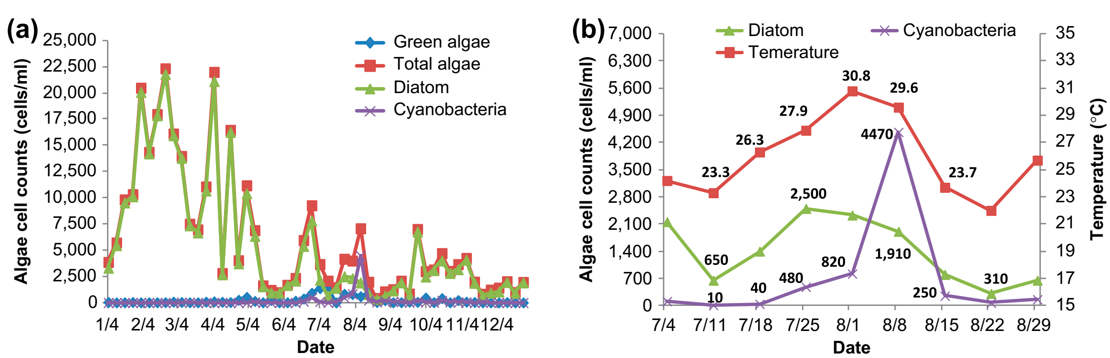

Fig. 3. Seasonal variation of dominant algae. (a) Variation of algae cell counts, and (b) variation of algae and temperature.

(from 23.3 to 27.9˚C), the diatom and cyanobacteria starts to augment at the same time. After the temperature steps up (27.9 ⇒ 29.6˚C), the cyanobacteria rapidly dominate the phytoplankton communities (480 ⇒ 4,470 cells/ml), while the diatoms were reduced (2,500 ⇒ 1,910 cells/ml). So, in the season of cyanobacterial blooms, the pre-chlorination treatment can cause the T&O compounds to destroy the cells of cyanobacteria. For removing the cyanobacteria, the treatment of pre-chlorination should be changed to that of inter-chlorination.

3.2. Analysis of particle-bound and dissolved geosmin

As it can be seen in Fig. 4(a), in July–August 2012, when large numbers of cyanobacteria flourish in a water body (an "algae bloom"), the cell count of cyanobacteria increases to 4,470.

At this time geosmin concentration increases to levels of 378 ng L^{-1}. These cyanobacteria synthesize geosmin during growth and these algal cells release or store these odorants depending on the growth phase and environmental factors. The geosmin released during the death and biodegradation of these cells [7]. These conditions lead to cyanobacterial blooms in the surface water resulting in significant geosmin production.

Here, special attention has to be paid to the analysis of dissolved as well as particle-bound geosmin compounds. The latter can be an important source of geosmin problems when particle-bound compounds are released from damaged algal cells during drinking water treatment.

As it can be seen in Fig. 4(b), the distribution difference of particle-bound and dissolved geosmin were relatively large. In the summer season, variation of particle-bound and dissolved geosmin was 8–311, 2–97 ng L^{-1}, respectively.

Fraction of particle-bound to total geosmin was measured as 67–86% (average value 77%). Some study shows that comparison of geosmin concentrations in raw water and filtered water showed that, on average, 90% of the geosmin could be associated with the particulate fraction [8]. So, the inter-chlorination treatment can reduce the number of particle-bound geosmin by using process of coagulation/flocculation.

3.3. Removal of geosmin with PAC and chlorination treatment

3.3.1. Removal efficiency with the low and medium geosmin levels

Pre-chlorination treatment has been conducted to reduce ammonia and organic matters from raw water. However, in case of outbreaks of cyanobacteria, pre-chlorination treatment increases the geosmin levels due to destroying the cells of cyanobacteria. So, to preserve the algae cell-destruction, the removal efficiency of geosmin was evaluated with types of chlorination.

The evaluation was classified with the low concentration of geosmin (<20 ng L^{-1}) and medium concentration of geosmin (20–80 ng L^{-1}). Fig. 5 illustrates the removal efficiency of geosmin with types of chlorination for the low and medium geosmin levels.

To reduce geosmin levels below threshold concentration (10 ng L^{-1}), the Amsa WTP was operated with two separated systems (1st WTP and 2nd WTP). Here, 1st WTP was operated with pre-chlorination (0.7 mg L^{-1} as Cl_2) and inter-chlorination (1.0 mg L^{-1} as Cl_2), and, 2nd WTP was operated with exclusive inter-chlorination (1.7 mg L^{-1} as Cl_2). The PAC dosage for low and medium geosmin levels was applied as 5 and 20 mg L^{-1}, respectively.

Fig. 5(a) and (b) shows the removal efficiency of low geosmin levels (5–15 ng L^{-1}) and medium geosmin levels (20–80 ng L^{-1}).

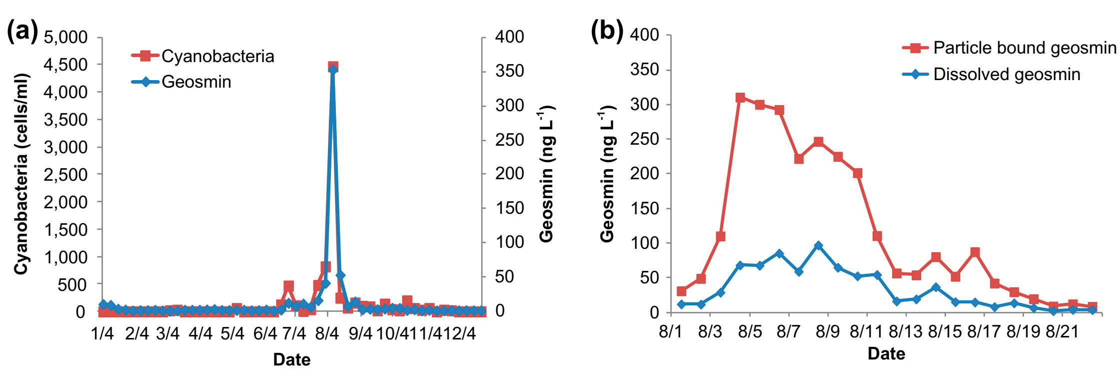

Fig. 4. Variation of particle-bound and dissolved geosmin. (a) Cyanobacteria and geosmin, and (b) particle-bound and dissolved geosmin.

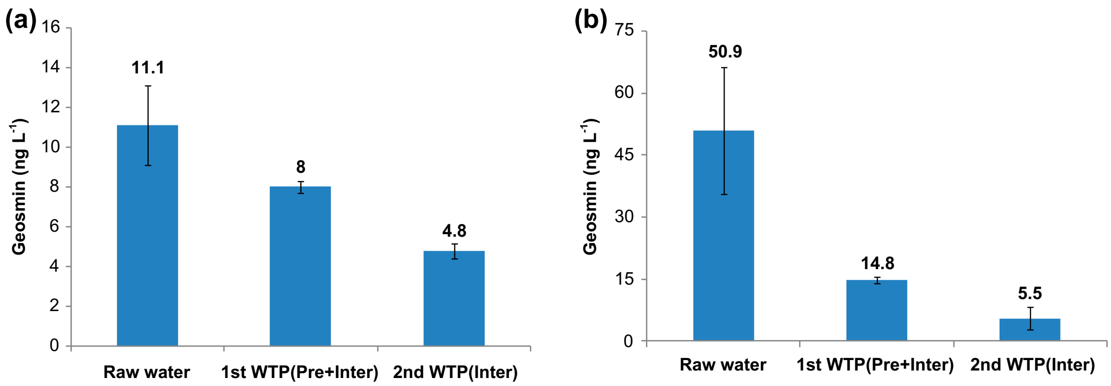

Fig. 5. Removal efficiency of geosmin with the low and medium geosmin levels. (a) Low concentration of geosmin and (b) medium concentration of geosmin.

As shown in Fig. 5(a), the average removal efficiency of geosmin in the 1st and 2nd WTP process was estimated at 26 and 54%, respectively. In Fig. 5(b), the average removal efficiency of geosmin in the 1st and 2nd WTP process was estimated as 69 and 88%, respectively. So, the exclusive inter-chlorination treatment (2nd WTP) was more effective than the treatment combined by pre-chlorination and inter-chlorination. It was proved to be reduced above two times at low geosmin levels and 1.2 times at medium geosmin levels.

Table 1
Amsa raw water quality (4–11 August)

Turbidity (NTU)	6.0–7.3
pH	7.3–7.4
Coagulant (mg L^{-1})	14–15

Note: No pre-chlorination treatment.

This reason was that pre-chlorination treatment makes levels of geosmin increase by destroying the cells of cyanobacteria. And, because geosmin was primarily bound to particles, removal of particle-bound geosmin would be more advantageous but care must be taken not to disrupt the cells [9]. Thus, the exclusive inter-chlorination treatment (2nd WTP) has a significant effect to remove the geosmin by not destroying the cells of cyanobacteria.

3.3.2. Effective treatment with the high geosmin levels

In Amsa WTP, the PAC dosage is possible to pump 30 mg L^{-1} due to limited capacity of PAC storage tank. In case of geosmin levels above 80 ng L^{-1} in summer season, it was very difficult to control the geosmin through PAC treatment only. Also, optimizing the PAC dosage was important since overdosing could result in excessive sludge production, reduced filter performance, and larger operating costs.

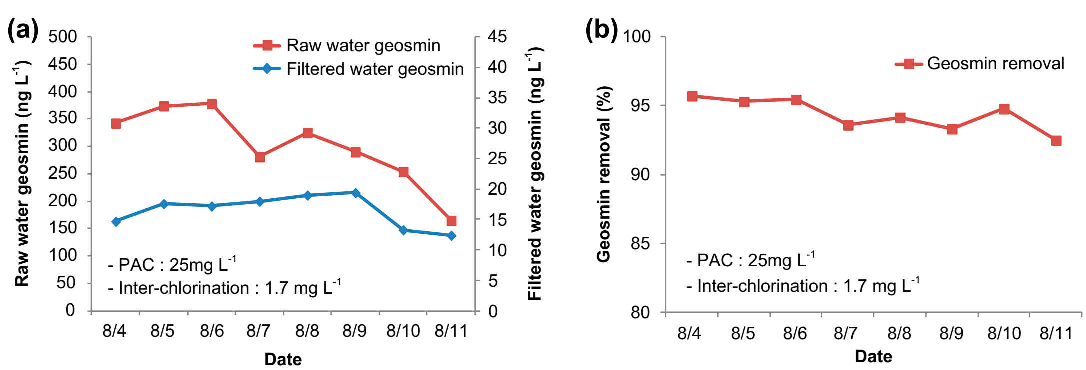

Fig. 6. Removal efficiency with the high geosmin levels. (a) Variation of geosmin during the study period and (b) removal efficiency of geosmin.

So, when high geosmin levels outbreaks, the combined treatment of PAC (25 mg L^{-1}) and inter-chlorination (1.7 mg L^{-1}) was applied to the water treatment process. In 4–11 August 2012, as shown in Fig. 4(a), the geosmin concentration increases to levels above 378 ng L^{-1} due to large numbers of cyanobacteria. At this time raw water quality is illustrated in Table 1.

As shown in Table 1, it consists of turbidity ranging from 6.0 to 7.3 nephelometric turbidity units, pH from 7.3 to 7.4, and coagulant from 14 to 15 mg L^{-1}.

Fig. 6(a) shows the variation of raw water geosmin (165–378 ng L^{-1}) and filtered water (12–19 ng L^{-1}). Fig. 6(b) shows the removal efficiency of geosmin.

In the season of cyanobacterial blooms (4–11, August), the removal efficiency of geosmin was estimated from 92.5 to 95.7% (average value 94.3%). Although geosmin of raw water was measured to be 378 ng L^{-1}, 18 times of Korean water quality standard (<20 ng L^{-1}), the filtered geosmin was treated as 19 ng L^{-1} safely. The exclusive inter-chlorination treatment with the high geosmin levels was more effective rather than that with the low and medium geosmin levels.

Thus, it is proved that the treatment of exclusive inter-chlorination with PAC was effective to reduce the geosmin in full scale WTP.

4. Conclusions

In this study, after the characteristics of algae and geosmin were analyzed, the removal efficiency of geosmin was evaluated by combined treatment of PAC and inter-chlorination in full scale. Total algae were occupied as diatom all year-round except for August. In the season of diatom blooms, it is necessary to apply pre-chlorination to intake station. The pre-chlorination treatment can prevent the filter-clogging by destroying the diatom cells. However, in the season of cyanobacterial blooms, the pre-chlorination treatment can cause the T&O compounds to destroy the cells of cyanobacteria. At this time the pre-chlorination treatment should be changed to that of inter-chlorination.

The geosmin can be released during the death and biodegradation of cyanobacteria. Special attention has to be paid to the analysis of dissolved geosmin as well as particle-bound geosmin compounds. Fraction of particle-bound to total geosmin was measured as 77%. So, the inter-chlorination treatment can reduce a number of particle-bound geosmin. The particle-bound geosmin can be easily removed by process of coagulation and sedimentation.

Also, the removal efficiency of geosmin was evaluated with the variation of geosmin levels. For low levels of geosmin, the average removal efficiency of 1st and 2nd WTP process was estimated as 26 and 54%, respectively. For medium geosmin levels, the average removal efficiency of 1st and 2nd WTP process was estimated as 69 and 88%, respectively. The exclusive inter-chlorination treatment (2nd WTP) was more effective than combined treatment (1st WTP) by pre-chlorination and inter-chlorination.

In the season of cyanobacterial blooms, the removal efficiency of geosmin was estimated from 92.5 to 95.7%. The exclusive inter-chlorination treatment with the high geosmin levels was more effective rather than treatment with low and medium geosmin concentration. Thus, we find out that the exclusive inter-chlorination has an important effect on removal of geosmin.

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