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Evaluation of organics reduction performance of the GAC filtration with regenerated carbons using the long-term operational data of drinking water treatment facilities

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ABSTRACT

Degradation of raw water quality promoted the introduction of GAC (granular activated carbon) filtration and ozonation into Korean drinking water treatment facilities in 1994 in order to cope with organic pollutants. This study focuses on the evaluation of organics reduction performance of the GAC filtration with regenerated carbons using long-term operational data. Three drinking water treatment facilities at Busan were selected for this purpose. It was found in this study that GAC filtration and ozonation helped these facilities to reduce the treated levels of DOC (dissolve organic carbon) as well as of 1,4-dioxane. Regeneration affected organics reduction performance of GAC filtration. The GAC filtration with regenerated carbons was found more effective for DOC reduction, while the filtration with virgin carbons was more effective for THM (trihalomethane) reduction. These results were related to the pore size change that occurred after the regeneration. The regeneration increased the meso pore volume but decreased the micro pore volume. The regeneration cycle was important for the decrease in the micro pore volume. The micro pore volume remained relatively unchanged after the first regeneration but was substantially decreased after the regeneration was repeated by more than second time.

Key words: 1,4-dioxane, dissolved organic carbon (DOC), granular activated carbon (GAC), pore size change, regeneration

HIGHLIGHT

- The GAC filtration with regenerated carbons was more effective for DOC reduction, and less effective for THM reduction than the filtration with virgin carbons. The long-term operational data of drinking water treatment facilities confirmed the reduction effectiveness of DOC and 1,4-dioxane by the GAC filtration and ozonation.

INTRODUCTION

The Korean water supply mainly focused on quantitative expansion from 1961 to 1990 in accordance with industrialization. As shown in Figure 1, the water supply capacity increased from 600,000 m^3/d in 1961 to 16,274,000 m^3/d in 1990 (KMOE 2008). The water supply coverage also increased from 17.1 to 78.4% during the same period (KMOE 2008). After 1990, the focus shifted from quantitative expansion to quality improvement. The media promoted such a shift. The media intensively reported water quality degradation. There was a report in 1989 that drinking water was contaminated by microbes and heavy metals (KMOE 2008). A series of reports then followed. There was a report of disinfection byproducts such as trihalomethane (THM) contamination in drinking water in 1990 (KMOE 2008). The Nakdong River, which was the main water source for the South-East region of the Republic of Korea, was seriously polluted (Kim & Yoon 2005). There was a phenol incident in 1991 (KMOE 2008), in which phenol discharged from an industry polluted the Nakdong river. There was a bad odor incident at the Nakdong river in 1992 (KMOE 2008).

Such successive incidents led to public distrust on drinking water quality. Public outcry promoted the Korean government to prepare countermeasures. One was the General Measures of Clean Water Supply (GMCWS) in 1989 and the other was the Water Quality Management Improvement Measures (WQMIM) in 1994. The GMCWS focused on an increase in wastewater treatment coverage (Joongangilbo 1989). It attempted to upgrade source water quality by building wastewater treatment facilities (Joongangilbo 1989). Nonetheless, raw water quality was not noticeably improved. The government, therefore, took

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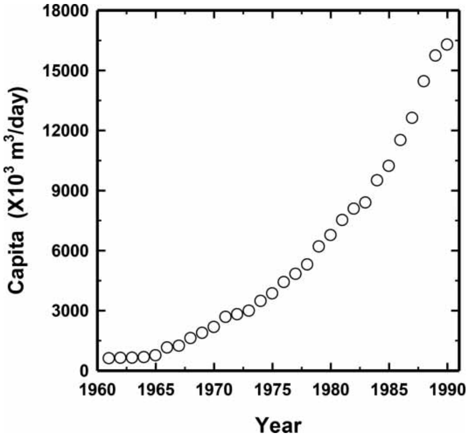

Figure 1 | Increase in the Korean water supply capacity of Korea from 1961 to 1990 (KMOE 2008).

another step. It was the WQMIM, at which the introduction of advanced water treatment was included (ME 1994). The government decided to introduce advanced water treatment into 21 drinking water treatment facilities (KMOE 2006). Detailed information on advanced water treatment introduced by the WQMIM is summarized in Table 1.

According to Table 1, advanced water treatment was mainly introduced into the facilities that relied upon the Nakdong river for their water sources (16 out of 21). In terms of the facility capacity, 94% was concentrated at the Nakdong river. The total facility capacity of advanced water treatment added was 5,446,500 m^3/d, and the Nakdong river portion was 5,139,500 m^3/d. Such concentration at the Nakdong river was caused by its water quality degradation. Increased discharge of untreated sewage and industrial wastewater into the Nakdong river in the 1990s resulted in serious water quality

Table 1 | Advanced water treatment introduced by the Water Quality Management Improvement Measures (WQMIM) in 1994 (KMOE 2006)

Water basin	Facility	Location	Capacity, 1,000 m^3/d	Process	Constructed
Nakdong river	Hwamyoung	Busan	600	GAC + Pre/post O_3	1994
	Myoungjang	Busan	277	GAC + Pre/post O_3	1999
	Deoksan	Busan	1,555	GAC + Pre/post O_3	2002
	Dooryu	Daegu	310	GAC + Post O_3	1997
	Maegok	Daegu	800	GAC + Post O_3	1998
	Moonsan	Daegu	200	GAC + Post O_3	–
	Hayang	Kyoungsan	10	PAC + GAC	1996
	Cheonsang	Ulsan	60	GAC + Post O_3	2002
	Hoeya	Ulsan	270	GAC + Post O_3	1999
	Samgye	Gimhae	165	GAC + Pre/post O_3	1999
	Myoungdong	Gimhae	210	GAC + Pre/post O_3	2003
	Beomeo	Yangsan	37.5	GAC + Post O_3	1999
	Woongsan	Yangsan	55	GAC + Post O_3	2001
	Seokdong	Jinhae	70	GAC + Pre/post O_3	1999
	Chilseo	Haman	400	GAC + Pre/post O_3	1998
	Bansong	Changwon	120	GAC	–
	Sub-total		5,139.5		
Han river	Dongduoocheon	Dongdoocheon	60	GAC	1998
	Moonsan	Paju	96	GAC + Post O_3	–
	Wonju 2	Wonju	85	GAC	2000
Keum river	Koonsan 2	Koonsan	38	PAC + GAC	1999
	Okryong	Gongju	28	GAC + Post O_3	1998
Total capacity			5,446.5		

GAC, granular activated carbon; PAC, powdered activated carbon; O_3, ozonation; –, under construction.

degradation of the Nakdong river, especially organic pollution. The biochemical oxygen demand (BOD) concentration of the Nakdong river exceeded 5 mg/L during those times (Kim & Yoon 2005). Subsequently, the government decided to introduce advanced water treatment since existing conventional treatments, such as coagulation, flocculation, sedimentation, and media filtration had limitations in coping with organic pollutants. According to Table 1, the main process of advanced water treatment was granular activated carbon (GAC) filtration and ozonation. GAC filtration combined with pre- and post-ozonation was the favorite choice of advanced water treatment in the WQMIM.

Almost 30 years have passed since advanced water treatment was introduced in 1994. The Busan Water Authority (BWA), which is responsible for drinking water treatment facilities in Busan, has been able to accumulate operational experience of GAC filtration including regeneration. It is, therefore, attempted in this study to evaluate the organics removal performance of both virgin and regenerated carbons using the long-term operational data of drinking water treatment facilities. Three Busan facilities were selected for this purpose. They were chosen because of their large production capacity and long operation period of GAC filtration. Their daily production capacity amounted to 2,432,000 m^3. The Hwamyoung facility has been operating the GAC filtration since 1994, the Myoungjang facility since 1999, and the Deoksan facility since 2002.

GAC filtration and ozonation at the Busan drinking water treatment facilities

The Busan drinking water treatment facilities possessed the same treatment processes (prechlorination, preozonation, coagulation, flocculation, sedimentation, media filtration, postozonation, GAC filtration, post chlorination). Information on advanced water treatment introduced at the Busan facilities is provided in Table 2. According to Table 2, the BWA installed the ozonation system even before the WQMIM announcement. The Hwamyoung facility has been operating the ozonation system since 1989, and the Deoksan #1 facility since 1995. The contact times of the ozonation system at these facilities were in the range of 2–12 min, and their ozone doses were in the range of 2.0–3.7 mg/L. These values generally corresponded to typical ranges reported in the literature. Kawamura (1991) recommended an ozone dosage of 1.5–3.0 mg/L, and a contact time of 5–15 min. The design and operation of GAC filtration were also in line with general practices. The GAC filter depth of the Busan facilities ranged from 3.0 to 3.5 m. The hydraulic loading ranged from 7 to 14 m/h, and the EBCT (empty bed contact time) ranged from 14 to 24 min. According to Roberts & Summers (1982), who surveyed design conditions for GAC filtration, the EBCT ranged from 5 to 24 min, the bed depth from 0.5 to 4 m, and the hydraulic loading from 2.6 to 17 m/h.

METHODS

DOC (dissolved organic carbon) concentration was measured by the TOC analyzer (Sievers, USA), and THM concentration was measured by the gas chromatography (Agilent, USA) with the electron capture detector. A predetermined amount of NaOCl was added into a sample of 70 mL so as to maintain a chlorine residual of 1 ppm for the trihalomethane formation potential (THMFP) measurement. After 24 h of reaction at 20 °C, THM concentration was measured. The $KMnO_4$

Table 2 | Information of advanced water treatment introduced at drinking water treatment facilities at Busan

		Deoksan plant				
Item	**Parameter**	**#1**	**#2**	**#3**	**Hwamyoung plant**	**Myoungjang plant**
GAC filter	Number	20	18	16	18	6
	Depth, m	3.0	3.0	3.0	3.35	3.5
	Volume, m^3	6,400	5,760	5,760	5,400	2,885
	EBCT, min	23	24	21	14	19
	Hydraulic loading, m/h	8	7	9	14	11
	Backwash	Air + water	Air + water	Air + water	Air + water	Air + water
	Carbon material	Coal	Coal	Coal	Coconut + coal → coal	Wood → coal
	Installed	2001	2001	2004	1994	1999
Ozonation	Contact time, min	5.8 (pre) 10 (post)	5.8 (pre) 10 (post)	5.1 (pre) 10 (post)	2 (pre) 8 (post)	6 (pre) 12 (post)
	Dose, mg/L	2–3.4	2–3.4	2–3.4	2.7	2.9 (pre) 3.7 (post)
	Installed	1992	1994	2004	1989	1998

EBCT, empty bed contact time.

consumption was measured in accordance with the Korean drinking water quality analytical methods (KMOE 2018). The concentration of 1,4-dioxane, which was purchased from Sigma-Aldrich, was measured by gas chromatography with mass spectrometry (Agilent, USA) in accordance with the Korean drinking water quality analytical methods (KMOE 2018).

Physical characteristics of GAC were measured in accordance with the Korean standards M (KMOE 2018), and the Korean drinking water quality analytical methods (KATS 2021). The pore volume of GAC was measured by the gas sorption analyzer (model iQ/MP-Xr, Quantachrome, USA). The 1,4-dioxane removal was examined by pilot-scale experiments. A pilot plant with a capacity of 100 m^3 per day was used for this purpose. The pilot plant processes consist of preozonation, rapid mixing, coagulation, sedimentation, media filtration, postozonation, and GAC filtration. The 1,4-dioxane at the predetermined concentration was fed into the preozonation.

RESULTS AND DISCUSSION

Organics reduction

Advanced water treatment helped reduce the treated organic level at the Busan drinking water treatment facilities. Results of the water quality improvement by advanced water treatment at the Busan facilities are summarized in Tables 3 and 4, as well as Figure 2. Bimonthly monitoring results of the DOC concentration by treatment processes from 1998 to 2020 are shown in Table 3 and Figure 2. The DOC levels were similar up to the conventional treatment of media filtration at the Hwamyoung and the Deoksan facilities, which took raw water directly from the Nakdong river. Average DOC concentrations of raw water were 3.12 and 3.13 mg/L at these facilities, respectively. These values were reduced to 2.04 and 2.05 mg/L after the media filtration, which indicated that both facilities could reduce the treated DOC concentrations by 35% through the conventional treatment. Further reduction of DOC was achieved through the GAC filtration, as shown in Figure 2. The average DOC concentration of the GAC filter effluent was 1.31 mg/L at the Deoksan facility, and 1.56 mg/L at the Hwamyoung facility. The GAC filtration helped these facilities to achieve additional 23 and 15% of DOC reduction, respectively. The influent DOC level at the Myoungjang facility, which took raw water from the reservoir under the influence of the Nakdong river, was lower than the other two facilities. The average DOC concentration of raw water was 2.68 mg/L. It was reduced to 1.58 mg/L after the media filtration, and then to 1.29 mg/L after the GAC filtration. Overall, the Busan facilities were able to reduce the treated DOC concentration by 35–41% through the conventional treatment, and by 50–58% after the GAC filtration.

The organics reduction performance was compared between the conventional and the advanced treatment at the Deoksan drinking water treatment facility using the operational data from 1996 to 2002. Such organics parameters as DOC, $KMnO_4$

Table 3 | DOC reduction after media filtration and GAC filtration over 1998–2020 at the Busan drinking water treatment facilities

Facility	Raw water	Media filtrated	GAC filtrated
Myoungjang	1.98–3.05 (2.68)[a]	1.13–2.10 (1.58)	0.87–1.70 (1.29)
Hwamyoung	2.60–3.87 (3.12)	1.68–3.01 (2.04)	1.21–2.48 (1.56)
Deoksan	2.58–3.91 (3.13)	1.76–2.52 (2.05)	0.83–1.90 (1.31)

[a] Values in the parenthesis indicate average values.

Table 4 | Comparison results of organics reduction between the conventional and the advanced treatment at the Deoksan drinking water treatment facility from 1996 to 2002

Description	Raw water	After the conventional treatment	After the advanced treatment
DOC, mg/L	2.70	1.35 (50%)[a]	0.95 (65%)
$KMnO_4$ consumption, mg/L	9.00	3.00 (67%)	1.25 (86%)
THMFP, μg/L	70	25 (64%)	10 (86%)
TTHM, μg/L	21[b]	32 (– 52%)	14 (33%)

[a] Values in the parenthesis indicate the reduction efficiencies.

[b] Pre-chlorinated water.

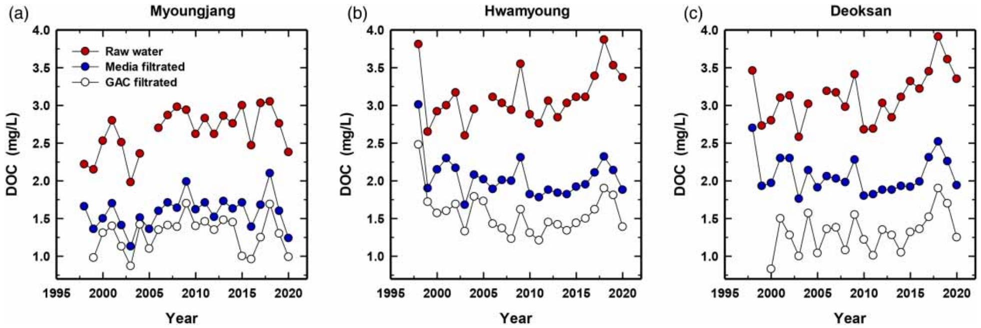

Figure 2 | Comparison of DOC concentration of raw water, media filtered, and GAC filtered over 1996–2020 at the Busan drinking water treatment facilities. Please refer to the online version of this paper to see this figure in colour: http://dx.doi.org/10.2166/aqua.2022.132.

consumption, THMFP, and total trihalomethane (TTHM) were used for the comparison. The comparison results are summarized in Table 4. The Deoksan facility has three units. The introduction timing of the advanced water treatment at these units was different. Unit #1 and #2 started operation of the advanced water treatment in March 2000, while unit #3 from April 2003. According to Table 4, the reduction efficiencies of $KMnO_4$ consumption and THMFP were greater than the DOC reduction efficiency. The $KMnO_4$ consumption level was reduced by 67% through the conventional treatment, and by 86% through the advanced treatment. On the other hand, the DOC reduction efficiencies were 50, and 65% after the conventional and the advanced treatment, respectively. THM formed at the presedimentation where prechlorination was provided. The TTHM concentration increased through the conventional treatment and then decreased as THM was adsorbed during the GAC filtration.

1,4-dioxane Reduction

The advanced water treatment also helped reduce the treated level of microorganic pollutants such as 1,4-doixane. It was first detected at the Hwamyoung and the Deoksan facilities in 2004. Its influent concentrations ranged in 2.5–5.1 μg/L at these facilities. Although these values were substantially lower than the World Health Organization drinking water quality guideline of 50 μg/L (WHO 2011), the BWA has decided to monitor this microorganic pollutant. They found that their facilities equipped with the advanced treatment successfully handled 1,4-dioxane. They were able to suppress the treated concentration of 1,4-dioxane as low as 1.0–2.4 μg/L.

The BWA also investigated the reduction performance of 1,4-dioxane by conventional treatment and advanced treatment using pilot plant experiments. They spiked 1,4-dioxane into their pilot plant so as to make its influent concentrations in the range of 2.6–36 μg/L. According to their pilot plant experimental results, the advanced treatment of ozonation and GAC filtration was effective for 1,4-dioxane reduction, but the conventional treatment was not. The preozonation reduced the 1,4-dioxane level by 49–70%, which remained unchanged through the media filtration. However, the GAC filtration following the postozonation further reduced the treated level of 1,4-dioxane as low as 0.5–1.3 μg/L. Overall reduction of 1,4-dioxane ranged from 83 to 97% at the pilot plant. This result indicated that the ozonation and GAC filtration was effective for 1,4-dioxane reduction.

GAC material

The Busan drinking water treatment facilities currently use only coal-based carbons although they initially selected GACs made of various materials of coal, coconut, and wood. The Myoungjang facility filled its GAC filters with wood-based carbons in 1998, which were replaced by coal-based carbons in 2015. Two different materials were introduced into the Hwamyoung facility, at which 14 GAC filters were filled with coconut-based carbons, and 4 filters with coal-based carbons in 1994. Coconut-based carbons were replaced by coal-based carbons in 2007. The Deoksan facility has been using coal-based carbons since 2001.

The carbon replacement at the Myoungjang facility was related to the GAC standards, shown in Table 5. The GAC standards of Korea (KMOE 2014) and Busan (Busan 2019) were shown in Table 5 together with the AWWA (American Water Works Association) standards (AWWA 2012). Wood-based carbons could not satisfy the apparent density requirement of the Busan standards. The apparent density of wood-based carbon (AWWA 2012) was reported in the range of 0.21–0.35 kg/m^3, which could not satisfy the Busan standards requirement of greater than 0.4 kg/m^3. Consequently, the Myoung-jang facility replaced wood-based carbons by coal-based carbons.

The Busan standards were stricter than the Korean standards for the adsorption characteristics such as the Iodine number and the methylene blue (MB) number. According to Table 5, the Korean standards require greater than 950 mg/g for the Iodine number, which can characterize the adsorptive capacity of GAC of smaller molecules (Alaya *et al.* 2000), while the Busan standards require 1,000 mg/g. Similarly, the Korean standards require the MB number, which can indicate the adsorption capacity of GAC of larger molecules (Alaya *et al.* 2000; Baçaoui *et al.* 2001; Nunes & Guerreiro 2011), greater than 150 mL/g. The Busan standards requires 200 mL/g. It is also noted in Table 5 that different parameters are used to characterize similar carbon properties. The Busan standards used the hardness number to characterize the carbon durability, while the AWWA standards used the abrasion number instead.

The carbon replacement at the Hwamyoung facility was related to the GAC adsorption capacity. The replacement was caused by the superior DOC reduction capacity of coal-based carbons. According to their operational data, coal-based carbons were more effective for DOC reduction than coconut-based carbons. The GAC filtration with coal-based carbons removed 5 kg of DOC per m^3 of carbons more than the filtration with coconut-based carbons, based on the bed volume of 19,000 (Ryu *et al.* 1995). However, both carbons showed similar adsorption capacity of chloroform. According to their operational data (Son *et al.* 2005), the chloroform breakthrough occurred at a bed volume of 10,929 with coal-based carbons. The corresponding bed volume was 10,210 with coconut-based carbons. However, the chloroform breakthrough occurred at the bed volume of 1,726 when wood-based carbons were used. Consequently, the Hwamyoung facility replaced coconut-based carbons by coal-based carbons.

Table 5 | Comparison among the granular activated carbon standards of Korea (KMOE 2014), Busan (Busan 2019), and the AWWA (AWWA 2012)

Parameter	Unit	Korean standards (KMOE 2014)	Busan standards (Busan 2019)	AWWA standards (AWWA 2012)
Apparent density	kg/m^3	–	>0.4	>0.2
Specific surface area	m^2/g	–	>1,060	>650
Iodine number	mg/g	>950	>1,000	>500
MB (Methylene blue) number	mL/g	>150	>200	–
Hardness number	%	–	>95	–
Abrasion number	%			>70
Ash content	%	<0.5	<0.5	<4
Moisture content	%	–	<9	<8
Effective size	mm	–	0.6–1.2	0.3–2.0
Uniformity coefficient	–	–	<2	<2.1
8 × 35 mesh	%	>95	>95	
Pore volume	cc/g	–	>0.6	–
Appearance	–	Black	Black	–
Experiment	–	Satisfied	Satisfied	–
pH	–	4.0–11.0	4.0–11.0	–
Chloride content	ppm	<0.5	<0.5	–
Arsenic content	ppm	<2	<2	–
Lead content	ppm	<10	<10	–
Cadmium content	ppm	<1	<1	–
Zinc content	ppm	<50	<50	–
Phenol content	ppm	<25	<25	–
ABS (Alkyl Benzene Sulfonate) content	ppm	<50	<50	–

Changes in the GAC characteristics with use

Characteristics of GAC changed with use. Changes in GAC characteristics such as the Iodine number, the MB number, the apparent density, and the ash content in accordance with the period of carbon use are summarized in Table 6. The Iodine number and the MB number of used carbons were smaller than those of virgin carbons, while the apparent density and ash content of used carbons were greater than those of virgin carbons. The Iodine number of used carbons decreased to 65% (687 mg/g) after use of 2 years, compared to the virgin carbon value of 1,050 mg/g, and to 55% (578 mg/g) after the use of 40 months, respectively. The decreasing extent of the MB number was less than that of the Iodine number. The MB number of used carbons decreased to 72% (152 mL/g) after use of 2 years, compared to the virgin carbon value of 212 mL/g and to 64% (135 mL/g) after 40 months. Unlike these adsorption characteristics, the apparent density and the ash content increased with an increasing period of carbon use. The apparent density and the ash content of used carbons increased to 110% and to 137% after use of 2 years, compared to the virgin carbon values, respectively. They increased further to 113 and 151% after the use of 40 months. An increase in the ash content also contributed to a decrease in the adsorption capacity of used carbons.

Regeneration conditions

The DOC reduction efficiency of GAC filtration deteriorated with time. Compared to the initial efficiency, the reduction efficiency deteriorated to 40–90% after the use of 10 months, to 12–25% after the use of 2 years, and to 4–10% after the use of 3 years. The BWA subsequently installed two multiple earth furnaces to regenerate used carbons in 2001. The capacity of one furnace was 12 m^3/d. The furnace capacity was determined, considering the GAC amount of 26,205 m^3 owned by the BWA, and the regeneration period of 3 years. The BWA conducted plant-scale experiments to determine the optimum regeneration conditions. They evaluated changes in the adsorption capacity of GAC, and its carbon recovery while monitoring the operational stability. The furnace temperature varied from 800 to 900 °C, the residence time from 25 to 90 min, and the injected steam quantity from 0.5 to 1.5 kg per kg of GAC during the experiments. Based on the experimental results, they determined the furnace temperature of 850 °C, the residence time of 35–40 min and the steam quantity of 0.75–1.0 kg per kg of GAC as the optimum regeneration conditions. Detailed information on the determination of the optimum regeneration conditions can be found elsewhere (Kim *et al.* 2005, 2015).

Effects of regeneration on the adsorption capacity of GAC

The BWA has regenerated used carbons up to six times. They found that the adsorption capacity of GAC gradually deteriorated with regenerations, as shown in Table 6. They examined changes in the Iodine number and the MB number of GAC at each regeneration. These adsorption characteristics of regenerated carbons were compared to those of virgin carbons, and the comparison results are summarized in Figure 3. Figure 3 also shows the THM breakthrough bed volumes (BVs). According to Figure 3, the adsorption capacity of GAC deteriorated with regenerations. The Iodine number of the RC-1 (the first regenerated carbons) decreased to 92% of that of virgin carbons, and the Iodine number of RC-2 (the second regenerated carbons) decreased to 76%. The decreasing extent of the MB number was less than that of the Iodine number. The MB number of RC-1 increased to 105% of that of virgin carbons, but that of RC-2 decreased to 90%. The regeneration deteriorated the THM adsorption capacity of GAC. The THM breakthrough of GAC filters came faster with regenerated carbons, compared to virgin carbons. The breakthrough BVs of RC-1 and RC-2 was 81, and 69% of that of virgin carbons.

Table 6 | Changes in the GAC characteristics with use (Kim *et al.* 2005)

GAC	Iodine number, mg/g	MB number, mL/g	Apparent density, g/cc	Ash content, %
Virgin carbons	1,050	212	0.48	7.3
Used carbons of 5 months	792 (75%)[a]	192 (91%)	0.50 (104%)	7.8 (107%)
Used carbons of 10 months	742 (71%)	160 (75%)	0.51 (106%)	8.5 (116%)
Used carbons of 24 months	687 (65%)	152 (72%)	0.53 (110%)	10 (137%)
Used carbons of 40 months	578 (55%)	135 (64%)	0.54 (113%)	11 (151%)

[a] Values in the parenthesis indicate the reduction efficiencies.

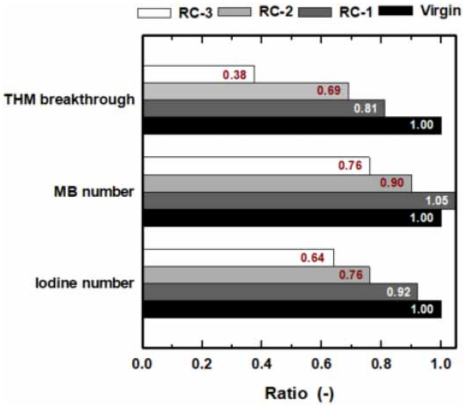

Figure 3 | The iodine and the MB number together with the THM breakthrough bed volume of the first, the second, and the third regenerated carbons and virgin carbons: RC-1, RC-2, and RC-3 indicate the first, the second, and the third regenerated carbons.

Effects of regeneration on the pore size change of GAC

The regeneration changed the pore volume distribution of GAC, as shown in Figure 4. Figure 4 shows pore volumes of virgin and regenerated carbons as a function of pore size. Carbons of the first, second, third, and fifth regenerations were included for the comparison in Figure 4. According to Figure 4, there was less micro pore (<2 nm) volume and more meso pore (2–5 nm) volume for regenerated carbons, compared to virgin carbons. This result means that the regeneration decreased the micro pore volume of GAC, but increased the meso pore volume. The regeneration cycle was also important for the change in the pore volume distribution of GAC. The regeneration substantially decreased the micro pore volume, but the extent of the decrease was minimal at least by the first regeneration. The decrease became significant after the second regeneration. The regeneration substantially increased the meso pore volume. The regeneration tended to increase larger pore volume with repetitions. According to Figure 4, RC-5 possessed more volume of the large pore (−4 nm), followed by RC-3.

Effects of regeneration on organics reduction performance of the GAC filtration

Effects of regeneration on organics reduction performance of the GAC filtration was different, depending upon the organics type. The calculated amounts of DOC and TTHM removed by one GAC filter (300 m^3) at the Hwamyoung drinking water treatment facility until its breakthrough was reached are summarized in Table 7. The calculation was performed based on the influent flowrates, influent and effluent concentrations, and time to breakthrough. Different breakthrough conditions were used for the calculation. The DOC breakthrough was considered to be reached when the DOC concentration of the

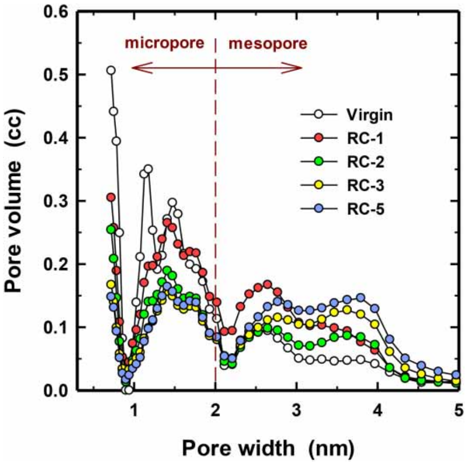

Figure 4 | Changes in the pore volume distribution of GAC with regenerations. Please refer to the online version of this paper to see this figure in colour: http://dx.doi.org/10.2166/aqua.2022.132.

Table 7 | Amounts of DOC and TTHM removed by one GAC filter until breakthrough at the Hwamyoung drinking water treatment facility

GAC	DOC, kg	TTHM, kg
Virgin carbons	2,654 (2,285–3,571)	52 (25–91)
RC-1	3,076 (2,673–3,409)	45 (16–65)
RC-2	3,771 (2,841–4,638)	22 (7–38)
RC-2 + 3	3,152 (2,618–4,146)	13 (4–24)
RC-3 + 4	4,656 (3,471–6,502)	17 (1–40)
RC-4 + 5	2,864 (2,816–2,912)	9 (4–15)
RC-1 + 2	2,615	26
RC-2 + 3 + 4	3,457	5
RC-2 + 5	2,887	19
RC-3 + 4 + 5	3,962	7
RC-1 + 4	3,136	24
RC-6	2,589	6

RC indicates regenerated carbons, and numbers indicate regenerated times. For example, RC-1 means the first regenerated carbons, and RC-2 + 3 means the second and the third regenerated carbons. Values in the parenthesis indicate the range measured, and the values outside the parenthesis indicate an average value.

GAC filter effluent reached 90% of its influent concentration. The TTHM breakthrough was considered to be reached when the filter effluent concentration of TTHM became identical as the influent concentration. It was noted in Table 7 that the GAC filtration with regenerated carbons generally removed more DOCs, but less TTHMs than the filtration with virgin carbons. on average, the GAC filtration with virgin carbons removed 2,654 kg of DOC, and 52 kg of TTHM until the breakthrough. The amount of DOC removed was increased to 3,076 kg, and the amount of TTHM was decreased to 45 kg for the GAC filtration with RC_1. The GAC filtration with repeatedly regenerated carbons generally extended the changes of the increase or the decrease. The GAC filtration with RC-2 removed 3,771 kg of DOCs, and 22 kg of TTHM. It was evident that the TTHM reduction performance of the GAC filtration deteriorated as the number of the regeneration was increased. The amount of TTHM removed was decreased to 6 kg when the GAC filter operation was conducted with RC-6. Unlike the TTHM reduction, the increasing trend of the DOC reduction was not evident as the number of regeneration was increased. The amounts of DOC removed by one GAC filter fluctuated after three regenerations.

The organics reduction performance of the GAC filtration was related to the pore volume distribution of GAC and the organics type. Large and small molecular organics are more easily adsorbed onto meso pores and micro pores of GAC, respectively. The Nakdong River, which the BWA relied on as their water source, was mostly composed of large molecular organics. It consisted of 42% of humic substances (HS), 26% of building blocks, and 27% of low molecular weight organics (Son *et al.* 2021). A relationship between HS removal and meso pore volume was reported previously. According to Gui *et al.* (2018), HS removal was closely related to the meso pore volume (3–24 nm). Golea *et al.* (2020) also reported similar results that the adsorption of DOC and disinfection byproduct precursor was closely related to the meso pores (5–10 nm). Therefore, the GAC filtration with regenerated carbons, which possess more meso pores and of less micro pores, was effective for DOC reduction, but less effective for THM reduction.

CONCLUSIONS

Organics reduction performance of the GAC filtration was evaluated using the long-term operational data in this study. This study focused on the GAC filtration with regenerated carbons. According to the operation data, GAC filtration together with ozonation helped reduce the treated levels of DOC as well as of 1,4-dioxane. The regeneration changed the pore size of GAC. The regeneration increased the meso pore volume, but decreased the micro pore volume. The regeneration cycle was important for the pore size change of GAC. The decrease in the micro pore volume was insignificant after the first regeneration, but was significant after the second regeneration. The GAC filtration with regenerated carbons was found more effective for DOC reduction, and less effective for THM reduction, compared to the filtration with virgin carbons. These results can provide insight into the GAC regeneration strategy. The purpose of the GAC filtration should be considered when to determine the regeneration strategy. When the main purpose of the GAC filtration is DOC reduction, the regeneration can be repeated as many as possible. The regeneration cycle may be extended to more than three or four times. On the other hand, when the GAC filtration is mainly used for the reduction of microorganic pollutants, the regeneration cycle should be minimized.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

REFERENCES

Alaya, M. N., Hourieh, M. A., Youssef, A. M. & El-Sejariah, F. 2000 Adsorption properties of activated carbons prepared from olive stones by chemical and physical activation. *Adsorpt. Sci. Technol.* **18**, 27–42.

AWWA 2012 AWWA standard: granular activated carbon, NASI/AWWA B604-122012.

Baçaoui, A., Yaacoubi, A., Dahbi, A., Bennouna, C., Phan Tan Luu, R., Maldonado-Hodar, F. J., Rivera-Utrilla, J. & Moreno-Castilla, C. 2001 Optimization of conditions for the preparation of activated carbons from olive-waste cakes. *Carbon* **39**, 425–432.

Busan 2019 GAC purchase specifications (in Korean).

Golea, D. M., Jarvis, P., Jefferson, B., Moore, G., Sutherland, S., Parsons, S. A. & Judd, S. J. 2020 Influence of granular activated carbon media properties on natural organic matter and disinfection by-product precursor removal from drinking water. *Water Res.* **174**, 115613.

Gui, H. H., Li, F. S., Wei, Y. F. & Yamada, T. 2018 Adsorption characteristics of natural organic matter on activated carbons with different pore size distribution. *Int. J. Environ. Sci. Tehcnol.* **15**, 1619–1628.

Joongangilbo 1989 *Drinking Water Pollution General Measures*. Available from: https://news.joins.com/article/2351703 (accessed June 24 2022). (in Korean).

Kawamura, S. 1991 *Integrated Design of Water Treatment Facilities*. John Wiley & Sons, Inc., New York.

Kim, S. H. & Yoon, J. 2005 Water quality improvement by advanced treatment processes of GAC filtration and ozonation over an eight-year period. *J. Water Suppl.: Res. Technol.-AQUA* **54** (2), 95–104.

Kim, H., Ryu, D., Kim, K., Na, Y., Ju, D., Ryu, B. & Ji, Y. 2005 Optimization of thermal reactivation for coal-based spent GAC used on the full-scale BAC process. *The Research Report of Water Quality Research Institute, Busan Water Authority* **9**, 102–108. (in Korean).

Kim, S. G., Son, H. J., Jung, J. M., Ryu, D. C. & Yoo, P. J. 2015 Evaluation of drinking water treatment efficiency according to regeneration temperatures of granular activated carbon (GAC). *J. Environ. Sci. Int.* **24** (9), 1163–1170. (in Korean).

Korean Agency for Technology and Standards (KATS) 2021 Test methods for activated carbon, KS M 1802. Available from: https://e-ks.kr/streamdocs/view/sd;streamdocsId=72059215016130445 (accessed June 24 2022). (in Korean).

Korean Ministry of Environment (KMOE) 2006 *Evaluation of Advanced Water Treatment Facilities*. Available from: http://www.me.go.kr/home/web/policy_data/read.do;jsessionid=Bot13nMLezrUHrbSYkTf5E+g.mehome1?pagerOffset=3720&maxPageItems=10&maxIndexPages=10&searchKey=&searchValue=&menuId=10259&orgCd=&condition.deleteYn=N&seq=2885 (accessed June 24 2022). (in Korean).

Korean Ministry of Environment (KMOE) 2008 *100 Years of Korean Water Supply*. pp. 169–207. (in Korean).

Korean Ministry of Environment (KMOE) 2014 *Standards and Specification for Drinking Water Chemicals*. (in Korean).

Korean Ministry of Environment (KMOE) 2018 *Korean Drinking Water Analytical Methods*. Available from: http://www.law.go.kr/DRF/lawService.do?OC=me_pr&target=admrul&ID=2100000204847&type=HTML&mobileYn= (accessed June 24 2022). (in Korean).

Maeil Economics (ME) 1994 *Water Quality Improvement Measures*. Available from: https://www.mk.co.kr/news/home/view/1994/01/2620/ (accessed June 24 2022). (in Korean).

Nunes, C. A. & Guerreiro, M. C. 2011 Estimation of surface area and pore volume of activated carbons by methylene blue and iodine numbers. *Quim. Nova* **34** (3), 472–476.

Roberts, P. V. & Summers, R. S. 1982 Performances of granular activated carbons for total organic carbon removal. *J. AWWA* **74** (2), 113–118.

Ryu, D. C., Kim, S. G., Lee, Y. S., Ryu, J. I. & Lee, S. H. 1995 The effect of water quality improvement by advanced water treatment. *The Research Report of Water Quality Research Institute, Busan Water Authority* **1**, 145–176. (in Korean).

Son, H. J., Roh, J. S., Kim, S. G., Bae, S. M. & Kang, L. S. 2005 Removal characteristics of chlorination disinfection by-products by activated carbon. *J. Korean Soc. Environ. Eng.* **27** (7), 762–770. (in Korean).

Son, H., Choi, S., An, B., Lee, H. & Yoom, H. S. 2021 Effect of changes in physical properties of granular activated carbon (GAC) in the adsorption of natural organic matter (NOM) with increasing the number of thermal regeneration: pore size and NOM molecular weight. *J. Korean Soc. Environ. Eng.* **43** (7), 537–546. (in Korean).

World Health Organization (WHO) 2011 *Guidelines for Drinking-Water Quality*, 4th edn, WHO, Geneva, p. 363.

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