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# Does Biological Activated Carbon Filtration Make Chlor(am)inated Drinking Water Safer

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**Abstract:** Biological activated carbon (BAC) filtration is an effective technology for the removal of natural organic matter. However, one potential drawback of BAC, especially old BAC, is that effluents can contain soluble microbial products released from the biofilm, which are recognized as more toxic nitrogenous DBPs (N-DBPs) precursors. So far, limited studies reported the risk of DBP formation potentials (FPs) increase caused by the microbial leakage of BAC. This study compared removal differences of DBP FPs between two BAC filters operated for 1 year and 8 years in a drinking water plant. The results showed that the total summed haloacetic acid FPs and trihalomethane FPs decreased by 34.31% from chlorination, and 55.01% of the total summed halogen acetonitrile FPs from chloramination were removed by the new BAC. However, Chlorinated haloacetonitriles FPs increased by 2.33% after old BAC filtration. To sum up, BAC filtration decreased most DBP FPs, but a potential risk regarding more toxic N-DBP FPs from old BAC should receive more attention.

**Keywords:** biological activated carbon; carbonaceous disinfection by-products; chloramination; chlorination; nitrogenous disinfection by-products



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## 1. Introduction

The disinfection of drinking water can effectively prevent the outbreak of cholera and other water-borne diseases through inactivating pathogenic microorganisms. Drinking water disinfection is considered to be one of the greatest achievements of public health in the 20th century [1,2], and currently, chlorine and chloramine are widely used for drinking water disinfection [3,4]. Compared with chlorine disinfection, chloramine disinfection reduces the formation of regulated carbonaceous disinfection by-products (C-DBPs), such as trihalomethane (THM) and haloacetic acid (HAA), but increases the formation of emerging unregulated nitrogenous DBPs (N-DBPs), such as halogen acetonitrile (HAN) [5–7]. Previous studies found that N-DBPs are usually more cytotoxic and genotoxic by several orders of magnitude than C-DBPs. Usually, dissolved organic carbon (DOC) and dissolved organic nitrogen (DON) are considered as important sources of C-DBPs and N-DBPs, respectively [8–10].

Recently, ozone-biological activated carbon ($O_3$-BAC) as an advanced treatment technology has been applied in drinking water treatment plants (DWTP) worldwide. It has an excellent performance in reducing disinfection by-product formation potential (DBP FPs) as it can remove DBP precursors such as natural organic matter to some degree by advanced oxidation, biodegradation, and adsorption [11–14]. However, some researchers reported that the biofilm on the surface of BAC has a risk to detach and release into the effluent [7,15,16]. For instance, Li, Wang, Xu, Xu, Wang and Xiao [17] found that DOC increased by 41% from the BAC influent to its effluent, and the BAC released a tyrosine-like protein and unrecognized high molecular weight compounds. Afterwards, microbes, soluble microbial products (SMPs), and extracellular polymeric substances (EPS) in BAC filters were found to release into the effluent [7], and usually, they were considered to be important precursors of N-DBPs [18–20].

For a long time, researchers tend to consider the BAC filter as an effective technology for the removal of natural organic matter, including some DBPs precursors, as well as algal toxins, ammonia, and trace level organics. However, recently, more and more evidence has indicated that BAC might produce the precursors of DBPs, especially N-DBPs, as stated earlier. So far, limited studies have reported the risk of an increase of DBP FPs caused by the microbial leakage of BAC. Shen, Tang, Wu and Chen [21] reported that SMPs were generated and leaked during the filtration of two BAC filters with running times of 3 and 6 years, resulting in an increase in HAA FPs after BAC treatment. However, little has been known if BAC treatment increase N-DBP FPs which should be more concerning as it is more toxic than C-DBPs, and biofilms composed of EPS, SMPs, and bacteria in BAC are important precursors of N-DBPs. Considering the old BAC filter removed fewer amino acids and polysaccharides but produced more amino sugar and proteins compared with the new filter [22], the authors hypothesized that the old BAC is more likely to increase the N-DBP yield than the new BAC. However, little study has investigated the effect of old BAC on N-DBP FPs, which may cause potential health risks to humans (e.g., genotoxicity, mutagenicity, and carcinogenicity). The effect of old BAC filtration on the C-DBP FPs and N-DBP FPs of effluents is a new crucial factor to be considered for the determination of the activated carbon replacement cycle in DWTP.

The purpose of this study was to evaluate the potential biological leakage risk of new and old BAC filters on the formation potential of two classes of C-DBPs (THM and HAA) and one class of N-DBPs (HAN) through the chlorine and the chloramine disinfection test.

## 2. Materials and Methods

### *2.1. Chemicals*

DBPs' standard solutions, including $THM_4$ (trichloromethane (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM) and tribromomethane (TBM)), $HAA_7$ (dichloroacetic acid (DCAA), trichloroacetic acid (TCAA), Bromochloroacetic acid (BCAA), dibromoacetic acid (DBAA), bromodichloroacetic acid (BDCAA), chlorodibromoacetic acid (CDBAA), tribromoacetic acid (TBAA)), $HAN_4$ (trichloroactetonitrile (TCAN), dichloroacetonitrile (DCAN), bromochloroacetonitrile (BCAN), and dibromoacetonitrile (DBAN)), as well as Methyl tert-butyl ether (MTBE, high performance liquid chromatography grade) were purchased from J&K Scientific Ltd. (Shanghai, China). Sodium hypochlorite (NaOCl), ammonium chloride ($NH_4Cl$), and all other chemicals were obtained from Sinopharm Chemical Reagent Co. (Shanghai, China). All solutions were prepared with ultrapure water produced from a Millipore Milli-Q Gradient water purification system (18 MΩ·cm, Billerica, MA, USA). All the chemicals used in this study were of analytical grade unless otherwise noted.

### *2.2. Experimental Procedures*

#### 2.2.1. Water Samples

To compare the differences in DBP FPs' removal from old and new BAC filters, the influent and the effluent of two BAC filters with different operation times were collected in January and October for subsequent DBP FP tests from a DWTP with the source water from Lake Taihu in Jiangsu Province, China. The main treatment processes include pre-ozonation, coagulation, precipitation, sand filtration, ozonation, BAC filtration, and chlorine disinfection. The activated carbon in many BAC filters can be used for 8–10 years [23–26]. The authors selected BAC filters with running times of 1 year and 8–9 years to represent new and old BAC filters in this study. Both the two BAC filters had been working normally in this DWTP. More details of the BAC filter can be found in Table 1. The old BAC filter may have a potential risk in terms of N-DBP FPs, and therefore should be paid attention to. Since the water matrix could impact the DBP FPs' removal efficiency by BAC, the new BAC filter in this study was considered as a reference to estimate old BAC performance. Once collected, the water samples were filtered through 0.45 μm microporous filter membranes

and then saved in the dark at 4 °C for less than 3 days before DBP FP tests. All the samples were in triplicate.

**Table 1.** Parameters of the BAC filters investigated in this study.

| GAC Bed Depth (m) | GAC Particle Size (mm) | Loading Density (g/dm$^3$) | Empty Bed Contact Time (min) | Backwash Time (min) |
|---|---|---|---|---|
| 2.1 | 0.88 | 488 | 20–30 | 7.0 |

2.2.2. Chlor(am)ine Disinfection By-Products Formation Potential Tests

DBP FP tests were performed in triplicate in 40 mL amber glass volumetric bottles under headspace-free conditions in the dark at controlled room temperature (25.0 ± 0.5 °C), based on the procedure reported in previous studies [27,28]. Briefly, a NaOCl stock solution with a concentration of 125 mg Cl2/L was prepared as the chlorine disinfectant. The fresh monochloramine (NH2Cl) solutions were prepared in advance following the procedures of Mitch [29]. In the DBP FP tests, the disinfection times of chlorine and chloramine were 24 h and 72 h, respectively. The chlorine and the chloramine demands, which were the initial dosages of chlorine and chloramine, were calculated according to Equations (1) and (2), respectively [27,28].

$$\text{Chlorine } (Cl_2) \text{ dosage} = 3 \times \text{DOC mg/L} + 7.6 \times NH_4{}^+\text{–N mg/L} + 10 \text{ mg/L} \quad (1)$$

$$NH_2Cl \text{ dosage} = 3 \times \text{DOC mg/L} \quad (2)$$

The water samples were collected from the bottles at the end of the experiment. Immediately, the DBPs were separated and extracted by MTBE via liquid–liquid extraction (LLE). Then, they were saved in the freezer for less than 24 h for the subsequent DBPs analysis.

*2.3. DBPs Measurement*

$THM_4$, $HAA_7$, and $HAN_4$ were chosen to be the target DBPs in this study because they were frequently detected in drinking water [30]. Detailed information on the analytical methods for DBPs is described in Table 2. They were measured using a gas chromatograph equipped with an electron capture detector (GC/ECD) (Clarus 680, PerkiElmer, PerkinElmer Co., Ltd., Waltham, MD, USA) based on the United States Environmental Protection Agency (USEPA) method 551.1 and 552.3.

**Table 2.** Analytical methods and conditions for DBP measurement.

| GC/ECD: | Clarus 680, PerkinElmer, USA | | |
|---|---|---|---|
| Columns: | Elite-5, 30 m × 0.25 mm ID, 0.25 μm film thickness | | |
| Carrier gas: | Nitrogen, constant flow at 3 mL per minute | | |
| Injection volume: | 1 μL | | |
| | Temperature programmes: | | |
| Vaporizing chamber: | 200 °C | | |
| | for $THM_4$ | for $HAA_7$ | for $HAN_4$ |
| GC column | Initial temperature at 37 °C for 3 min, then 10 °C per minute to 80 °C and hold for 2 min and finally 20 °C per minute to 220 °C and hold for 1 min. | Initial temperature at 40 °C for 7 min, then 2.5 °C per minute to 65 °C and 5 °C per minute to 85 °C and hold for 1 min finally 20 °C per minute to 210 °C and hold for 5 min. | Initial temperature at 30 °C for 10 min, then 17 °C per minute to 72 °C and hold for 1 min and finally 40 °C per minute to 200 °C and hold for 2 min. |
| Detector: | 300 °C | | |

### 2.4. Analytical Methods

DOC and total dissolved nitrogen (TDN) in the influent and effluent of the BAC filters were measured by a total organic carbon analyzer (Jena multi C/N 2100, Analytik Jena AG, Jena, German) after prefiltration through a 0.45 μm membrane. UV absorbance at 254 nm was detected by a spectrometer (UV752, Shanghai Jingke Co., Ltd., Shanghai, China).

## 3. Results

### 3.1. Water Characteristics

The characteristics of the water samples used in this study are provided in Table 3. The influent DOC and TDN were both slightly higher in January than in October: 4.02 mg/L DOC and 1.49 mg/L TDN for January, and 3.81 mg/L DOC and 1.36 mg/L TDN for October. The effluent DOC was slightly lower after the new BAC filtration than the old BAC filtration, indicating the new BAC filter removed more DOC than the old. In addition, the effluent DOC was higher after the old BAC filter than the value supplied for the influent DOC in January. The relatively low $SUVA_{254}$ of 0.68–0.96 indicates that the organic matter was more hydrophilic and non-aromatic in the influent and the effluent of the BAC [31,32]. There was no difference in TDN between the influent and the effluent of the BAC.

**Table 3.** Basic water quality parameters of water samples ($n$ = 3).

| Sampling Time | Water Sample | DOC (mg/L) | $SUVA_{254}$ | TDN (mg/L) |
|---|---|---|---|---|
| January | Influent | 4.02 ± 0.09 | 0.82 | 1.49 ± 0.01 |
| | Effluent of new BAC | 3.01 ± 0.15 | 0.86 | 1.53 ± 0.04 |
| | Effluent of old BAC | 4.09 ± 0.21 | 0.68 | 1.50 ± 0.02 |
| October | Influent | 3.81 ± 0.26 | 0.93 | 1.36 ± 0.06 |
| | Effluent of new BAC | 3.35 ± 0.24 | 0.67 | 1.62 ± 0.18 |
| | Effluent of old BAC | 3.51 ± 0.26 | 0.96 | 1.37 ± 0.06 |

### 3.2. The Formation of Chlorine Disinfection By-Products

Table 4 presents the formation potential of THMs, HAAs, and HANs during the chlorination of the influent and the effluent of the old and new BAC filters. The concentrations of THM FPs, HAA FPs, and HAN FPs decreased from 58.55 ± 35.86 μg/L, 421.84 ± 18.49 μg/L, and 64.85 ± 42.50 μg/L in the influent to 48.50 ± 32.94 μg/L, 208.55 μg/L, and 60.36 ± 28.23 μg/L in the effluent of the new BAC filter, and 55.26 ± 37.40 μg/L, 373.95 ± 4.23 μg/L, and 65.93 ± 42.14 μg/L in the effluent of the old BAC filter. The total summed C-DBP FPs were decreased by 34.31 ± 14.50% and 11.56 ± 2.33% by the new BAC and the old one, respectively, indicating that the new BAC removed more C-DBP precursors than the old one, which is in line with previous studies [7,33]. The total summed HAN FPs levels increased by 0.35 ± 22.23 % for the new BAC, and 2.33 ± 2.07% for the old one, which was mainly caused by the increase in DCAN FP, TCAN FP, and BCAN FP. The coconut shell-activated carbon-quartz sand biofilter combined gas-water backwashing is an appropriate choice for the removal of HAN FPs [34,35].

It can be clearly observed in Figure 1 that the new BAC filter evidently decreased THM FPs, HAA FPs, and HAN FPs, while the old BAC presented relatively poor removal ability in DBP FPs, especially DCAN, TCAA, TCAN, BDCM, and TBAA. Why did the old BAC filter present a poor removal ability? On the one hand, GAC usually has a limited adsorption lifetime. For instance, Tan, Lin, Jiang, Dong, Chen and Zhou [36] reported that after two years of BAC operation, the GAC adsorption capacity decreased significantly. Therefore, it can be speculated that in this study, the adsorption capacity of the old BAC was exhausted. On the other hand, Zheng, Lin and Chen [15] reported that the old BAC attached more microorganisms, but showed low biological activity and more SMP yield, which could result in the increase in N-DBPs precursors in the effluent. The results in Table 3 showed that more DOC representing the precursors of C-DBPs (THM FPs and HAA

FPs) were removed by the new BAC filter, which is consistent with the results of the DBP FPs, as shown in Figure 1.

**Table 4.** Formation potential of THMs, HAAs, and HANs during chlorination of the influent and the effluent of old and new BAC filters. ($n$ = 3).

| DBP Category | Sampling Time | DBP Species | Influent Concentration (μg/L) | Effluent Concentration in New BAC (μg/L) | Effluent Concentration in Old BAC (μg/L) |
|---|---|---|---|---|---|
| THMs | Jan. | TCM | 60.81 ± 0.40 | 50.06 ± 2.52 | 57.80 ± 1.45 |
| | | BDCM | 18.37 ± 2.68 | 17.92 ± 1.18 | 19.41 ± 0.27 |
| | | DBCM | 4.73 ± 0.43 | 3.80 ± 0.17 | 4.50 ± 0.07 |
| | | TBM | Not Detected | Not Detected | Not Detected |
| | Oct. | TCM | 22.71 ± 1.08 | 16.51 ± 0.65 | 19.40 ± 0.70 |
| | | BDCM | 6.81 ± 0.25 | 5.59 ± 0.01 | 6.29 ± 0.49 |
| | | DBCM | 3.33 ± 0.31 | 2.87 ± 0.10 | 2.83 ± 0.14 |
| | | TBM | 0.34 ± 0.04 | 0.30 ± 0.01 | 0.27 ± 0.02 |
| | | Total $_{average}$ | 58.55 ± 35.86 | 48.50 ± 32.94 | 55.26 ± 37.40 |
| HANs | Jan. | DCAN | 64.25 ± 2.51 | 53.09 ± 2.78 | 64.94 ± 8.65 |
| | | TCAN | 7.82 ± 0.19 | 7.46 ± 0.18 | 7.97 ± 0.31 |
| | | BCAN | 7.35 ± 0.66 | 6.35 ± 0.21 | 8.61 ± 0.56 |
| | | DBAN | 15.49 ± 0.70 | 13.41 ± 0.36 | 14.21 ± 0.79 |
| | Oct. | DCAN | 1.18 * | 1.54 ± 0.11 | 1.93 ± 0.17 |
| | | TCAN | 19.41 ± 0.01 | 16.82 ± 1.57 | 19.06 ± 0.98 |
| | | BCAN | 6.68 ± 0.13 | 7.18 ± 0.36 | 7.22 ± 0.60 |
| | | DBAN | 6.90 * | 14.82 ± 0.43 | 7.91 ± 0.89 |
| | | Total $_{average}$ | 62.86 ± 42.50 | 60.36 ± 28.23 | 65.93 ± 42.14 |
| HAAs | Jan. | DCAA | 128.76 ± 8.10 | 73.20 ± 9.57 | 95.57 ± 4.62 |
| | | TCAA | 73.83 ± 6.50 | 38.53 ± 3.93 | 73.55 ± 2.16 |
| | | BCAA | 36.02 ± 1.79 | 16.57 ± 2.88 | 25.29 ± 0.64 |
| | | DBAA | 23.24 ± 1.16 | 14.96 ± 3.44 | 19.94 ± 0.83 |
| | | BDCAA | 28.30 ± 0.25 | 16.69 ± 3.89 | 21.40 ± 1.08 |
| | | DBCAA | 71.36 ± 2.17 | 19.54 ± 5.58 | 73.98 ± 1.31 |
| | | TBAA | 60.31 ± 1.87 | 29.02 ± 1.44 | 64.20 ± 1.44 |
| | | Total | 421.84 ± 18.49 | 208.55 ± 46.13 | 373.95 ± 4.23 |

Note: * Refers to only a single data.

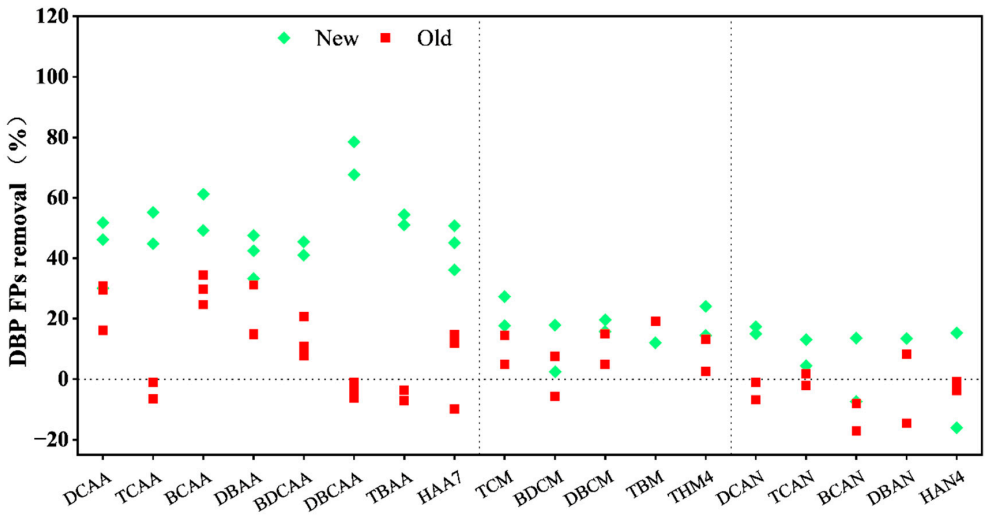


**Figure 1.** The removal of formation potential of THMs, HAAs, and HANs during the chlorination of old and new BAC filters. Scattered point of HAA FPs represents a single sample and other scattered point represents the average of three replicate samples.

### *3.3. The Formation of Chloramine Disinfection By-Products*

Table 5 presents the formation potential of THMs, HAAs, and HANs during the chloramination of the influent and the effluent of the old and new BAC filters. The concentrations of THM FPs, HAA FPs, and HAN FPs decreased from 8.03 ± 4.65 μg/L, 86.80 μg/L, and 70.51 ± 8.74 μg/L in the influent to 4.47 ± 2.41 μg/L, 76.09 μg/L, and 30.43 ± 16.87 μg/L in the effluent of the new BAC filter, and 6.34 ± 3.80 μg/L, 82.10 μg/L, and 49.30 ± 0.55 μg/L in the effluent of the old BAC filter. The total summed C-DBP FPs decreased by 31.25 ± 14.74% and 14.99 ± 11.37% by the new BAC and the old BAC, and the total summed N-DBP FPs decreased by 55.01 ± 29.60% and 29.50 ± 7.94% by the new BAC and the old BAC. It indicates that the BAC filter removed more N-DBP precursors than C-DBP precursors for the subsequent chloramination.

**Table 5.** Formation potential of THMs, HAAs, and HANs during chloramination of the influent and the effluent of old and new BAC filters. ($n$ = 3).

| DBP Category | Sampling Time | DBP Species | Influent Concentration (μg/L) | Effluent Concentration in Newew BAC (μg/L) | Effluent Concentration in Od BAC (μg/L) |
|---|---|---|---|---|---|
| **THMs** | **Jan.** | TCM | 3.02 ± 0.51 | 1.97 ± 0.10 | 2.31 ± 0.17 |
| | | BDCM | 4.65 ± 0.24 | 2.25 ± 0.09 | 3.60 ± 0.11 |
| | | DBCM | 3.65 ± 0.09 | 1.95 ± 0.02 | 3.11 ± 0.04 |
| | | TBM | Not Detected | Not Detected | Not Detected |
| | **Oct.** | TCM | 1.50 ± 0.01 | 0.51 ± 0.27 | 0.84 ± 0.05 |
| | | BDCM | 1.48 ± 0.03 | 0.96 ± 0.06 | 1.33 ± 0.03 |
| | | DBCM | 1.54 ± 0.01 | 1.12 ± 0.03 | 1.31 ± 0.05 |
| | | TBM | 0.22 ± 0.01 | 0.18 ± 0.003 | 0.17 ± 0.002 |
| | | Total $_{average}$ | 8.03 ± 4.65 | 4.47 ± 2.41 | 6.34 ± 3.80 |
| **HANs** | **Jan.** | DCAN | 34.37 ± 3.18 | 12.17 ± 1.11 | 27.69 ± 6.49 |
| | | TCAN | 6.62 ± 0.50 | Not Detected | 6.03 ± 0.41 |
| | | BCAN | 23.24 ± 1.81 | 4.36 ± 0.53 | 10.79 ± 1.68 |
| | | DBAN | 12.46 ± 1.31 | 1.97 ± 0.41 | 5.17 ± 0.82 |
| | **Oct.** | DCAN | 0.63 * | 0.24 * | 0.41 * |
| | | TCAN | 52.32 ± 2.06 | 32.90 ± 3.83 | 38.71 ± 0.21 |
| | | BCAN | 7.18 ± 0.19 | 6.30 ± 0.50 | 6.65 ± 0.45 |
| | | DBAN | 4.21 ± 0.92 | 2.92 ± 0.04 | 3.14 ± 0.10 |
| | | Total $_{average}$ | 70.51 ± 8.74 | 30.43 ± 16.87 | 49.30 ± 0.55 |
| **HAAs** | **Jan.** | DCAA | 24.48 ± 2.73 | 20.01 ± 0.15 | 27.70 ± 1.06 |
| | | TCAA | 12.27 ± 0.63 | 13.21 ± 0.38 | 15.18 ± 0.37 |
| | | BCAA | 9.61 ± 0.32 | 5.99 ± 0.03 | 7.21 ± 0.14 |
| | | DBAA | 4.72 ± 0.16 | 2.79 ± 0.25 | 3.14 ± 0.01 |
| | | BDCAA | 8.48 ± 0.18 | 7.70 ± 0.13 | 8.46 ± 0.41 |
| | | DBCAA | 9.77 ± 0.25 | 9.57 ± 0.07 | 9.86 ± 0.13 |
| | | TBAA | 5.41 ± 0.48 | 5.06 ± 0.20 | 5.28 ± 0.45 |
| | | Total | 77.73 ± 2.81 | 64.33 ± 5.41 | 73.84 ± 1.77 |

Note: * Refers to only a single sample.

It is clearly observed in Figure 2 that the new BAC filter evidently decreased the THM FPs, HAA FPs, and HAN FPs, while the old BAC presented relatively poor removal ability in DBP FPs, especially TCAA, BDCAA, DBCAA, and TBAA. The results showed that the THM, HAA, and HAN yield from chloramination were far less than that from chlorination, indicating that chloramine as a disinfectant may reduce the chemical risk regarding DBP formation.

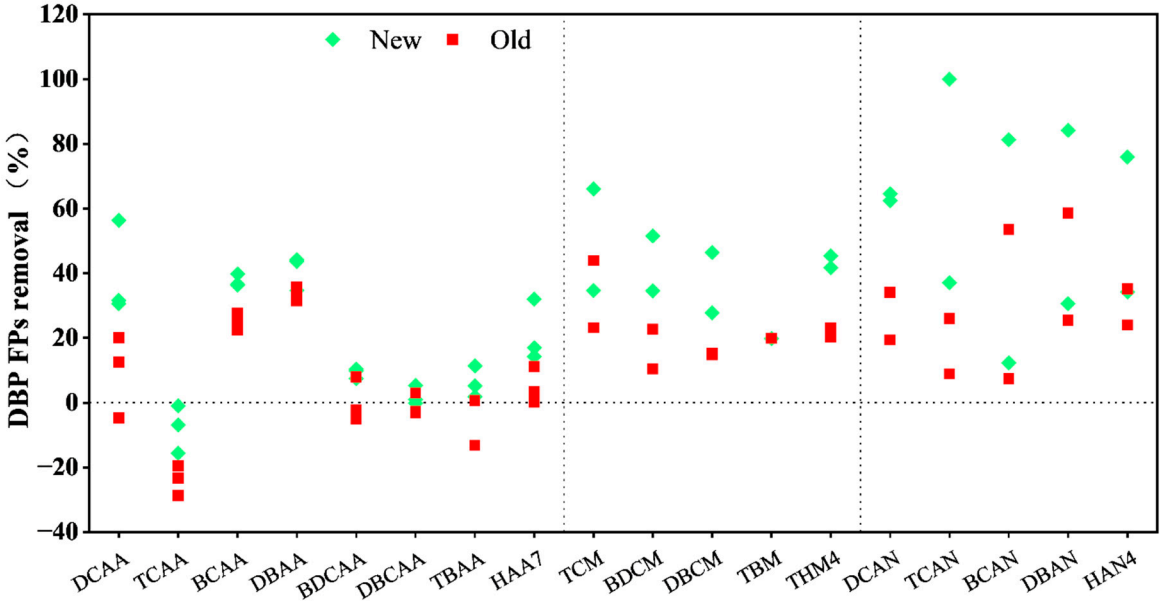


**Figure 2.** The removal of formation potential of THMs, HAAs, and HANs during the chloramination of old and new BAC filters. The scattered point of HAA FPs represent a single sample, and the other scattered point represents the average of three replicate samples.

## 4. Conclusions

The new BAC filter removed more DOC than the old one, indicating that more precursors of THMs and HAAs were removed by the new BAC. For chlorinated DBPs, the removal efficiencies of THM FPs and HAA FPs were 12.59% and 50.56% by the new BAC and 7.92% and 11.35% by the old BAC. Differently, HAN FPs increased by 0.35% and 2.33% after the filtration of the new BAC and old BAC, respectively. The results proved that biological leakage might occur and result in an increase in N-DBP FPs. For chloraminated DBPs, almost all chloraminated DBP FPs decreased after BAC filtration while the new BAC performed better than the old BAC as well.

In conclusion, the new BAC filter performed better than the old one for the removal of DBP FPs during both chlorination and chloramination. Not only the regulated parameters but also the DBP FPs, especially N-DBP FPs, should be considered for managers and regulators to determine the activated carbon replacement cycle when chlorine disinfection follows BAC filtration.

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