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Re-assessing ICR GAC Treatment Study Database: Effect of Bromide on DBP Formation

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Abstract

While granular active carbon (GAC) can effectively remove disinfection byproduct (DBP) precursors, its use has raised concerns over increased formation of some brominated DBP species in treated water following postchlorination, especially for waters with high bromide concentrations. The Information Collection Rule Treatment Study Database contains results of the most extensive GAC studies ever conducted nationwide. Data were analyzed to assess the extent of DBP speciation changes and overall reduction of brominated DBPs by GAC to gain new insights of the bromide effect. Results showed that formation of three brominated trihalomethanes (collectively, Br-THM3) varied greatly depending on TOC removal and bromide concentrations. Low TOC concentrations in GAC effluents resulted in greatly reduced Br-THM3 formation, except for a few cases where Br-THM3 formation increased. GAC followed by chloramination were likely to better control Br-THM3 formation for waters with high TOC and high bromide. Lastly, the chlorine demand reduction by GAC was quantified.

Keywords

GAC; brominated DBPs; total organic carbon; bromide

INTRODUCTION

Granular activated carbon (GAC) adsorption has long been used to remove synthetic organic chemicals, taste and odor compounds, and natural organic matter (NOM) from drinking water supplies (Chowdhury et al., 2013). Recently, many water utilities have installed or are considering installing GAC and/or other advanced technologies (e.g., ozone, membrane filtration) as a protective or mitigation measure against source water contamination, such as algal toxins (e.g., Westrick et al., 2010), chemical spills, and per- and polyfluoroalkyl substances (e.g., Dickenson & Higgins, 2016). Because it often exists in a much higher concentration (in mg/L) than trace organics (in µg/L or ng/L) in water, NOM can interfere with the adsorption of trace organics and be removed by GAC simultaneously. Removal of

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NOM reduces the formation of most disinfection byproducts (DBPs) in treated water following postchlorination (Chiu et al., 2012; Singer, 1994). Lower organic matter in water also reduces disinfectant dosage and helps maintain disinfectant residuals in distribution systems (Roth & Cornwell, 2018).

Since the discovery of trihalomethanes (THMs) in drinking water in 1974, more than 600 DBPs have been identified and many others are still unknown (Richardson et al., 2011; 2007). Epidemiological studies have suggested consistent association between exposure to THMs in chlorinated water and bladder cancer (Cantor et al., 2010; Villanueva et al., 2007; 2004) and less consistent associations with various adverse reproductive and developmental effects (USEPA, 2016). To reduce human exposure to DBPs in chlorinated water, the US Environmental Protection Agency (USEPA) established maximum contaminant levels (MCLs) for the sum concentrations of four THMs (THM4) and five haloacetic acids (HAA5) under the Stage 1 and Stage 2 Disinfectants and Disinfection Byproducts (D/DBP) Rules (USEPA, 2006; 1998). Individual species for these groups are not regulated due to the complexity of such regulation (USEPA, 1994). During chlorination, the presence of bromide leads to the formation of brominated DBPs (Br-DBPs), which have been shown to be more cyto- and genotoxic than their chlorinated analogs (Wagner & Plewa, 2017; Richardson et al., 2007). Ozonation of water containing bromide forms bromate (Haag & Hoigne, 1983; Gunten & Hoigne, 1994), which is also a regulated DBP. Recently concerns have been raised over the increasing concentrations of Br-DBPs as a result of increased bromide levels in drinking water sources (Good & VanBriesen, 2016; McTigue et al., 2014; States et al., 2013) and potential public health risks (Sawade et al., 2016; Regli et al., 2015; Richardson et al., 2007).

Extensive research has been conducted on the effect of GAC on DBP formation in the presence of bromide in water (Jiang et al., 2018; Stanford et al., 2017; Krasner et al., 2016; Chiu et al., 2012; Summers et al., 1993; Symons et al., 1993; Amy et al., 1991). GAC removes dissolved organic matter (often measured as total organic carbon, TOC), but has little effect on bromide. As a result, the Br/TOC ratio in the GAC effluent is higher than its influent, thus causing a shift toward formation of more brominated species. Previous studies have shown that GAC adsorption resulted in a higher bromine incorporation into DBPs and an increased formation of some Br-DBP species, such as bromoform (Symons et al., 1993) and dibromoacetonitrile (Krasner et al., 2016). In a recent study, GAC has been shown to effectively remove precursors for THM4, HAA5, HAA9, and nitrosamines in the presence of elevated bromide (Stanford et al., 2017). Despite of increased formation of some Br-DBP species and based on the predicted toxicity values of over 70 DBPs measured in that study, the overall cyto- and genotoxicity of the water was reduced by GAC (Stanford et al., 2017). However, these studies were limited to a few source waters and focused on the behavior of individual Br-DBP species.

Under the 1996 Information Collection Rule (ICR), a national assessment of GAC and membrane treatment for DBP precursor removal was conducted by public water systems that met USEPA-established criteria (Allgeier, et al., 1998; USEPA, 1996a). The participating systems included surface water systems (and ground water systems under the direct influence of surface water) serving 100,000 or more people and having 4 mg/L of TOC in

source water, and ground water systems serving 50,000 or more people and having 2 mg/L of TOC in finished water. Results from 99 treatment studies, 63 with GAC and 36 with membrane filtration, are stored in the ICR Treatment Study Database (ICRTSD) (Hooper & Allgeier, 2002). Due to high levels of TOC and co-occurring bromide in waters, some participating plants would face challenges of meeting the DBP standards under consideration at that time. For the GAC studies, either rapid small-scale column tests (RSSCT) or pilot-scale tests were conducted to develop breakthrough curves of TOC and DBP formation. Each breakthrough curve was fitted with a logistic function model that can predict the GAC bed life and effluent water quality for any given value of a breakthrough parameter (USEPA, 2000).

The wealth of information in the ICRTSD has led to various analyses on DBP precursor removal. Hooper and Allgeier (2002) evaluated the feasibility of GAC and membrane technologies in meeting regulatory options and helped establish best available technologies under the Stage 2 D/DBP Rule. Bond and Digiano (2004) examined key factors affecting the TOC breakthrough, e.g., feed water quality, GAC particle size, and empty bed contact time (EBCT), and developed predictive models for GAC service life. However, neither study reported the Br-DBP data.

The main objective of this work is to examine the extent of reductions in brominated THM formation by GAC under a broad range of water quality and treatment conditions to gain new insights on the bromide effect. A standardized dataset was created from the ICRTSD by setting the TOC breakthrough values at 1 and 2 mg/L, respectively, to provide a common basis for evaluating Br-DBP formation as a function of bromide concentrations. While USEPA has no maximum regulatory concentration limit for TOC, under the Stage 1 D/DBP Rule conventional treatment and softening plants having source water TOC > 2 mg/L are required to meet specific TOC removal percentages (USEPA, 1998). The 1- and 2-mg/L TOC values were chosen to cover a range of GAC operation for DBP control. In light of an increased interest in maintaining disinfectant residuals in distribution systems (Roth & Cornwell, 2018), this paper also evaluated the reduction of chlorine demand by GAC.

METHODS AND MATERIALS

Data source and queries.

The main data source is the ICRTSD available on the USEPA website (<https://www.epa.gov/dwsixyearreview/supplemental-data-six-year-review-3>). Two queries were conducted in the GAC module based on a single contactor TOC breakthrough of 1 and 2 mg/L, respectively. For each query, water quality parameters (e.g., TOC, ultraviolet absorbance at 254 nm (UV254), bromide, THM species, and chlorine demand) for the GAC influent and effluent were exported for all test runs. Specific ultraviolet absorbance (TSUVA) was calculated from UV254 divided by the TOC concentration. In addition, full-scale plant water quality data and treatment and disinfection practice information were downloaded from the ICRTSD, including annual average concentrations of TOC, bromide, UV254, THM4 and HAA5 for plant source and finished water. GAC run length data for single and blended contactors were exported for all test runs and described in the Supplemental Information (SI).

In the GAC studies, DBP formation was analyzed with simulated distribution system (SDS) tests using free chlorine. The SDS tests were intended to simulate average conditions (e.g., incubation time, pH, temperature, and chlorine residual) in the distribution system at the time of the particular study run, which varied seasonally and between plants (see SI Table S1). The significant variations in the SDS conditions (e.g., incubation time ranged from 4 to 207 hours) present a challenge for a direct comparison of DBP formation in all 33 studies. However, GAC influent and effluent samples from the same test run were analyzed under the same SDS conditions, which allowed “Delta” SDS DBP to be calculated.

In this paper, only THM4 data were analyzed because they contain all four bromine- and chlorine-substituted THM species and can capture any shift among the four species. In contrast, HAA5 or HAA6 data were not analyzed because they neglect several bromine-substituted HAA species and would not capture the shift among all nine species. Br-THM3 (the sum of bromodichloromethane, dibromochloromethane, and bromoform) is used as a surrogate to broadly represent the mixture of Br-DBPs in chlorinated water (see the SI for supporting rationale). Recognizing that the toxicities tend to increase with the number of bromide ions contained in the THM molecule (Wagner & Plewa, 2017; USEPA, 2006), this paper also includes individual THM species data in the SI.

Data filtering, cleaning and processing.

Starting with the original data queried from the ICRSTD, a reduced, consistent dataset was produced for analysis. The steps for data filtering and cleaning are described in the SI. The final dataset used for analysis contained records for 33 surface water studies conducted in 14 states, including 21 RSSCT studies (147 test runs) and 12 pilot studies (22 test runs). Each of the 33 studies was conducted by one ICR plant, except one plant that conducted two studies – one RSSCT and one pilot study. See details in the SI.

For each study (designated by a unique Treatment Study Identification Number, or TSID), annual average values for each parameter (e.g., TOC, UV254, bromide, and SDS-THMs) were calculated based on data from multiple test runs. Data from individual test runs reflect seasonal variations and the effect of EBCTs (10 vs. 20 min) on the GAC performance, which were not the focus of this work. This “averaging” approach was consistent with that previously used to characterize different GAC options for compliance with the MCLs under the Stage 2 D/DBP Rule (Hooper & Allgeier, 2002). For THM4 and Br-THM3, concentrations of individual species were first summed up at the test run level, and then averaged by TSID. Runs with missing data for any of the four THM species were eliminated. The minimum reporting levels required for the ICR treatment studies were 20 µg/L for bromide, 0.5 mg/L for TOC and 1 µg/L for each THM species (USEPA, 1996b). In this paper, any bromide values below 20 µg/L were replaced with 20 µg/L for consistency with the protocol used to synthesize and analyze the ICR full dataset (Wysock et al., 2002). In the ICRTSD, all “non-detect” values for each THM species were replaced with zero. Average concentrations were calculated and used to develop graphs and statistical summaries in this paper.

Based on the types of primary and residual disinfectants used in the full-scale plants conducting GAC studies (Table 1), these plants were separated into two groups: one group

with seven free chlorine (FC) plants and another with 25 chloramine (CM) plants. The FC plants used free chlorine and/or chlorine dioxide for primary disinfection and free chlorine for residual disinfection whereas the CM plants used chloramines as residual disinfectants and various types of primary disinfectants. Regardless of the plant disinfection practice, free chlorine was used in all the SDS-DBP tests. Table 2 summarizes water quality data of 33 GAC studies, including full-scale plant source and finished water, GAC influent and GAC effluent at 1- and 2-mg/L TOC.

Data for all 33 GAC studies were plotted, grouped by sequential IDs, to allow adequate resolution of individual observations. Each sequential ID consisted of an FC or CM plant code, a sequential number and average GAC influent bromide concentration (in µg/L) from low to high. For example, the study identified as CM2-24 is the second “chloramine” plant with an average influent bromide concentration of 24 µg/L. Note that the bromide concentration is equivalent to the Br/TOC ratio (at 1-mg/L TOC) or twice the ratio (at 2-mg/L TOC).

Calculations of Bromine Incorporation Factor (BIF).

BIF is a term used to indicate the degree of bromine substitution in DBPs. The BIF for THMs is defined as follows (Symons et al., 1993):

$$\text{BIF} = \frac{0[\text{CHCl}_3] + 1[\text{CHCl}_2\text{Br}] + 2[\text{CHClBr}_2] + 3[\text{CHBr}_3]}{[\text{CHCl}_3] + [\text{CHCl}_2\text{Br}] + [\text{CHClBr}_2] + [\text{CHBr}_3]}$$

where each concentration is on a molar basis.

After calculating a BIF value for each sample, the results were averaged by sample point and TSID, resulting in a yearly average BIF for the GAC influent and effluent for each TSID.

RESULTS AND DISCUSSION

Full-scale ICR plant water quality.

Water quality data of source and finished waters and distribution systems for all 500 ICR plants have been characterized extensively (McGuire & Hotelling, 2002). See the SI (Figures S2 to S4) for more details on TOC and bromide occurrence from the 1998 ICR dataset and a more recent 2011 dataset (no bromide data) for surface water systems serving greater than 100,000 people. This paper focuses on a subset of the ICR plants that conducted GAC studies due to their high source water TOC. For these 32 plants, average source water TOC concentrations ranged from 3.2 to 9.1 mg/L with a median of 5.2 mg/L (see Table 2). This median value was higher than that in the 1998 ICR full dataset for surface water systems and the 2011 ICR datasets (2.7 and 2.3 mg/L, respectively, see Figure S2). Cumulative distributions of source water TOC indicate that TOC concentrations of these 32 plants comprised the upper 20th to 30th percentiles of both the 1998 and 2011 datasets, representing some of the highest TOC waters in the nation (see Figure S2). A similar pattern is also seen in finished water TOC concentrations, which ranged from 2.1 to 4.8 mg/L (average 3.4 mg/L) across all 32 plants (see Table 2 and Figure S3). Bromide levels in these source waters varied widely from < 20 to 1,110 µg/L (median of 55 µg/L), exhibiting a

similar range but almost twice as high the median value as compared to the 1998 dataset (55 vs. 30 µg/L) (see Figure S4).

Of the 32 ICR plants, 21 (66%) employed conventional treatment and 11 (34%) a treatment train consisting of softening. As shown in Table 2, percent TOC removal by full-scale treatment processes varied widely among the CM plants (2 to 68%; average 32%) whereas the FC plants removed 44 to 58% of TOC (average 51%). The reduction in UV254 exceeded the corresponding reduction in TOC for both CM plants (2 to 86%; average 48%) and FC plants (49 to 86%; average 71%) due to the preferential removal of hydrophobic, UV-absorbing substances over TOC (Liang & Singer, 2003). As a result, the average value of TSUVA, an indicator of water susceptibility to enhanced coagulation (Archer & Singer, 2006), decreased from 2.9 L/mg-m in raw water to 1.9 L/mg-m in finished water.

THM4 and HAA5 concentrations in the full-scale plant distribution systems also varied widely and reflected the conditions before promulgation of the Stage 1 D/DBP Rule in December 1998. The FC plants had 45 to 105 µg/L of THM4 (average 71 µg/L) and 22 to 59 µg/L of HAA5 (average 42 µg/L) in their distribution systems. The CM plants had 2.6 to 102 µg/L of THM4 (average 48 µg/L) and 3.1 to 70 µg/L of HAA5 (average 30 µg/L) in their distribution systems (see Figure S5 of the SI).

GAC influent.

Water feeding the GAC study was usually collected from a plant before the addition of any disinfectants or oxidants. When a chlorine-based oxidant was added at a point that precluded such sampling, either raw or partially treated water was collected and used for the GAC study after full-scale treatment processes were simulated in the bench-scale treatment. The range and average TOC concentrations, UV254 and TSUVA values for GAC influent were comparable to those in the plant finished water (Table 2), indicating that GAC pretreatments adequately simulated the TOC removal at the full-scale plants. Pretreatment such as enhanced coagulation and softening can lower the TOC concentration in GAC influent, thereby extending the GAC bed life (Bond & Digiano, 2004). However, these processes can also remove the more adsorbable hydrophobic NOM fraction and decrease the adsorbability of NOM, as suggested by the lower TSUVA value – an index for aromaticity/hydrophobicity (Huang & Reckhow, 2012) (see Figure S9). Other pretreatment such as ozonation can transform precursor molecules into by-products that are less reactive with chlorine (Amy et al., 1991). The effects of different types of pretreatment on organic precursors, which may ultimately influence the Br-DBP formation, is beyond the scope of this paper.

Figure 1 presents TOC and bromide concentrations in the GAC influent for all 33 studies, showing no clear trend of co-occurrence. This lack of co-occurrence is consistent with what was reported for the ICR plant source waters (McGuire & Hotaling, 2002). Six CM plants on the righthand side of Figure 1 showed both high TOC (> 4.0 mg/L) and high bromide levels (> 150 µg/L), representing very challenging waters for compliance with the DBP MCLs. Not surprisingly, these plants used chloramines or chlorine dioxide (one combined with ozone) for primary disinfection. Overall, the CM plants had slightly higher average TOC concentrations (3.6 vs. 2.9 mg/L) but significantly higher bromide concentrations (<20 to 1,490 µg/L; average 192 µg/L) than the FC plants (<20 to 80 µg/L; average 34 µg/L).

(note: 1,490 µg/L is higher than the maximum bromide level of 1,110 µg/L in raw water presumably due to different sampling time.) These data indicate that plants with relatively high TOC and high bromide concentrations often used alternative disinfectants to minimize DBP formation.

The stacked column plot in Figure 2 shows SDS chloroform (bottom bar), Br-THM3 (top bar) and THM4 formation (sum of the two) in the GAC influent across all 33 studies. These values represent a baseline of DBP formation prior to GAC adsorption. Compared with the measured THM4 concentrations in the distribution system (2.6 to 105 µg/L; average 53 µg/L), SDS THM4 formation in the GAC influent showed a higher range and average concentration (29 to 234 µg/L; average 104 µg/L). This was particularly true for the CM plants, due, in part, to the use of free chlorine in the SDS tests. For the FC plants, since the same type of disinfectant was used in the plants and in the SDS tests, the difference between measured THM4 and SDS THM4 formation was largely attributed to the predictability of SDS tests. For example, some plants reported that SDS conditions in their treatment studies were intended to provide conservative estimates of the DBP formation potential.

Figure 2 also shows a clear trend that, as bromide concentrations increased, particularly among the CM plants, the dominating species shifted from chloroform to Br-THM3 with increasingly higher bromide concentrations (see Figure S6 for THM speciation). At the highest bromide concentration, nearly all THM4 was composed of brominated species with most being bromoform and dibromochloromethane. The GAC influent BIF varied from 0.1 to 2.8 (average 0.8) for all studies and displayed a generally increasing trend with higher bromide levels in Figure 3.

GAC effluent at 1-mg/L TOC breakthrough.

The stacked column graph in Figure 4a shows SDS chloroform, Br-THM3 and THM4 formation in the GAC effluent in all studies when the TOC concentration reached 1 mg/L (GAC1). Figure 4a also plots, on the second y-axis, average THM4 concentrations in the full-scale plant distribution systems (note: distribution system DBP speciation data are not available in the ICRTSD). All the FC plants had < 40 µg/L THM4 formation in GAC1, which were 60 to 90% lower than the respective THM4 concentrations measured in the distribution system. Approximately 80% of the CM plants had < 40 µg/L THM4 formation in GAC1; none exceeded 80 µg/L. Due to the use of alternative disinfectants, some CM plants had very low THM4 concentrations in their distribution systems (e.g., as low as 2.6 µg/L for CM5-32). The data indicate that THM4 levels may increase if these CM plants were to use GAC and free chlorine disinfection in lieu of using alternative disinfectants alone.

Similar to Figure 2, Br-THM3 formation in GAC1 (Figure 4a) also exhibited an increasing trend as bromide levels increased (note: the bromide level is the same as the Br/TOC ratio because the denominator is one). The BIF values varied from 0.4 to 3.0 (average 1.5) for the GAC1 across all studies (Table 2). Previous research has found that the degree of bromine substitution into DBPs depends on the ratio of bromide and chlorine dose, organic precursor availability and reactivity, as well as disinfection conditions (Hua & Reckhow, 2012; Sohn et al., 2006; Liang & Singer, 2003; Symons et al., 1993). These relationships were not further

examined in the present analysis. At a low TOC concentration and a high Br/TOC ratio, formation of brominated THMs may dictate the speciation due to fast reaction kinetics (Summers et al., 1993). As shown in Figure 3, BIF values consistently increased after GAC, as reported in previous studies (e.g., Krasner et al., 2016; Chiu et al., 2012). These researchers have postulated that GAC adsorption preferentially removes hydrophobic fraction of organic matter, leaving hydrophilic fraction and bromide in treated water. Upon chlorination, the higher Br/TOC ratio and hydrophilic, more reactive organic matter in treated water would result in higher bromine incorporation. In the present work, the average TSUVA value further decreased from 1.9 L/mg-m in the GAC influent to 1.1 L/mg-m in GAC1, demonstrating that GAC continued to remove hydrophobic fraction of organic matter from the low-TSUVA water which is less amenable to enhanced coagulation. See SI Figure S9 for more detailed characterizations.

Differences between Figures 2 and 4a, or delta chloroform, Br-THM3 and THM4 formation between GAC influent and GAC1, are shown in Figure 4b (left-hand y-axis). Percent TOC removal is plotted on the right-hand y-axis. Across all 33 studies, GAC removed 45 to 81% (average 70%) of TOC and 54 to 94% (average 81%) of UV254 from its influent to reach the 1-mg/L TOC target. As a result, THM4 formation decreased by 42 to 96% (average 72%) and Br-THM3 formation by zero to 94% (average 42%) (Table 2). None of the studies showed an increase in Br-THM3 formation after GAC. The extent of Br-THM3 reduction appears to be driven by the bromide level (other factors such as SDS conditions and the nature of NOM were not examined in this work.). At low to moderate bromide levels (e.g., <150 µg/L), GAC adsorption primarily reduced chloroform formation and had little or no effect on Br-THM3 formation. As the bromide level increased to 150 µg/L and higher, GAC adsorption primarily reduced Br-THM3 formation. Therefore, GAC decreased Br-THM3 formation in all the cases at the 1-mg/L TOC breakthrough even though the BIF values increased. However, formation of bromoform sometimes increased in the effluent (see Figure S7 and SI for more details), indicating that GAC was not always able to control the formation of bromoform during postchlorination as previously reported (Symons et al., 1993; Summers et al., 1993).

GAC effluent at 2-mg/L TOC breakthrough.

Compared to GAC1, THM4 formation in the 2-mg/L TOC effluent (GAC2) was significantly higher as more organic precursors were available to form DBPs (Figure 5a). The median and average THM4 concentrations almost doubled compared to GAC1; six plants exceeded 80 µg/L. Br-THM3 formation was also higher, ranging from 5.0 to 119 µg/L (average 38 µg/L) (Table 2). The BIF values ranged from 0.3 to 2.9 (average 1.2) for GAC2, still showing an increase from the GAC influent, but to a less extent compared to the case for GAC1 (see Figure 3). GAC2 had an average TSUVA value of 1.3 L/mg-m, which is slightly higher than GAC1 (1.1 L/mg-m) but considerably lower than GAC influent (1.9 L/mg-m).

For all the FC plants, THM4 formation in GAC2 was 9 to 77% lower than the respective measured THM4 concentration in the plant distribution system. Mixed results are shown in the CM plants, which, again, was due to the use of alternative disinfectants in the CM plants. If these plants were to use GAC and free chlorine disinfection (in lieu of their continued use

of alternative disinfectants), their THM levels may increase, particularly for those having relatively high bromide concentrations ($150 \mu\text{g/L}$). Therefore, there is a tradeoff between potential increases in THM levels and avoiding the unintended consequence associated with the use of alternative disinfectants (e.g., nitrification associated with the use of chloramines).

Figure 5b shows delta chloroform, Br-THM3 and THM4 formation between the GAC influent and GAC2 across all 33 studies. GAC removed 17 to 62% of TOC (average 41%) from its influent to reach the 2-mg/L target (note: one study, CM4-29, was excluded because its influent TOC concentration, e.g., 1.8 mg/L, was lower than the target of 2 mg/L, resulting in a negative removal.) The UV254 reduction ranged from 27 to 77% (average 57%). As a result, THM4 formation decreased by 23 to 71% (average 46%). Br-THM3 formation, however, increased in six studies by 0.3, 0.4, 1.4, 3.5, 6.0 and 11.4 $\mu\text{g/L}$, respectively. The first three Br-THM3 increases appeared to be negligible. The last three represented 29%, 40% and 41% of increases compared to the respective GAC influent. Table S2 of the SI presents water quality data for these three studies, all of which occurred at FC plants (e.g., FC3-43, FC6-20, and FC7-61). TOC concentrations in the GAC influent were 3.5, 2.4, and 2.7 mg/L, respectively. The influent bromide levels at these six plants were relatively low with the highest being 61 $\mu\text{g/L}$. For waters with moderate to high bromide concentrations, Br-THM3 formation did not increase, but bromoform increased for most of these plants (e.g., in 12 studies – see Figure S8).

For the aforementioned three studies, Br-THM3 formation decreased in GAC1 but increased in GAC2 compared to the GAC influent. Assuming that the GAC influent quality, bromide concentration, and SDS conditions were held constant, the difference between GAC1 and GAC2 is the TOC level and NOM composition (or reactivity). Based on the TOC concentrations in GAC influent, GAC1 represents 29%, 42%, and 37% of bed exhaustion in terms of TOC adsorption, respectively, all before the 50% bed exhaustion. Conversely, GAC2 represents 57%, 83%, and 74% of bed exhaustion, respectively, all after the 50% bed exhaustion. Therefore, this analysis suggests that Br-THM3 formation, with exception of bromoform, can be effectively controlled in the early stage of GAC operation (50% exhaustion). As the GAC adsorber is operated beyond the 50% exhaustion point towards 70% to 80% exhaustion, there is a likelihood that Br-THM3 formation may increase in the GAC effluent.

It is well established that GAC adsorption can lead to a higher BIF in treated water (Stanford et al., 2017; Chiu et al., 2012; Summers et al., 1993; Symons et al., 1993; Amy et al., 1991). However, this analysis shows that the TOC removal by GAC may offset the increase in BIF, especially before 50% of bed exhaustion is reached when considering Br-THM3 as a surrogate metric. However, since Br-THM3 is an aggregate metric which may sometimes decrease in concentration while the bromoform concentration increases, its indicator value may be compromised for those situations.

Impact of GAC treatment on chlorine demand.

Chlorine demand is influenced by the amount of organic DBP precursors present in water (Gang et al., 2002). It is also dependent on iron and manganese concentrations in water and other water quality parameters, such as pH and temperature, as well as the initial chlorine

dose (Roth & Cornwell, 2018; Katz, 1986). Although SDS conditions varied between sites, all the GAC influent and effluent samples from the same test run were analyzed under the same SDS conditions. Therefore, the test condition factor is eliminated when “delta” chlorine demand or reduction in chlorine demand between paired GAC influent and effluent is evaluated for individual test runs.

The stacked column plot in Figure 6 shows the chlorine demand of the GAC influent (entire column), GAC1 (bottom bar), and GAC2 (sum of bottom and middle bars) across all 33 studies. Chlorine demand in the GAC influent ranged from 1.4 to 5.6 mg/L as Cl₂ (average 3.0 mg/L as Cl₂). The delta chlorine demand between the GAC influent and GAC1 ranged from 0.6 to 3.8 mg/L as Cl₂ (average 1.9 mg/L as Cl₂), representing a reduction of 27 to 82% (average 63%). The delta chlorine demand between the GAC influent and GAC2 ranged from 0.2 to 3.6 mg/L as Cl₂ (average 1.4 mg/L as Cl₂), representing a reduction of 6 to 67% (average 46%). The lower chlorine demand in GAC effluent could decrease the applied chlorine dose and make it easier to maintain sufficient disinfectant residuals in distribution systems.

The middle bar represents the delta chlorine demand between GAC2 and GAC1, ranging from zero to 1.0 mg/L as Cl₂ (average 0.5 mg/L as Cl₂). As the TOC breakthrough continued from 1 to 2 mg/L as Cl₂, chlorine demand in the GAC effluent had increased by zero to 1.0 mg/L as Cl₂ (average 0.5 mg/L as Cl₂).

SUMMARY

Application of GAC to reduce TOC and thereby DBP formation can lead to a higher proportion of Br-DBPs in treated water upon postchlorination. Analysis of Br-DBP data from the ICRTSD GAC studies revealed that GAC generally lowered THM4 and Br-THM3 formation in treated water even though the BIF values increased. Formation of Br-THM3 varied depending on TOC removal by GAC and bromide concentrations. All 33 GAC studies showed decreases in the average Br-THM3 formation at 1-mg/L TOC whereas about 90% of the studies showed decreases and 10% showed slight increases in the average Br-THM3 formation at 2-mg/L TOC. However, it should be noted that the THM levels achieved for some CM plants, especially those at the 2-mg/L TOC breakthrough, may not be as low as those achieved in the full-scale plants that employed alternative disinfectants for the DBP control. The formation of bromoform increased in the GAC effluent, indicating that GAC was not always able to control the formation of bromoform during postchlorination as previously reported.

The extent of Br-THM3 reduction appears to be driven by the bromide level when TOC was held constant. At low to moderately high bromide levels (e.g., <10 to 150 µg/L), GAC primarily reduced chloroform formation and had little or no effect on Br-THM3 formation. As the bromide level increased to 150 µg/L and higher, GAC significantly reduced Br-THM3 formation because the shift to brominated-THMs was more than offset by the decreasing availability of organic precursors. Interestingly, the most significant reduction in Br-THM3 formation was observed in GAC influent containing high TOC (> 4 mg/L) and high bromide concentrations (> 150 µg/L). This was somewhat unexpected because previous

work suggested that GAC and other separation processes may not effectively control Br-THMs for waters containing significant amounts of bromide (e.g., Summers et al., 1993). However, from a practical standpoint, these utilities may continue to struggle with DBP compliance even after the GAC installation. The use of chloramines may be warranted to further reduce THM formation in distribution systems.

Removing TOC to 1 or 2 mg/L by GAC also effectively lowered the chlorine demand of water, achieving an average of 63% and 46% reduction, respectively. This reduction would decrease the amount of chlorine or chloramines needed for maintaining disinfectant residuals in distribution systems. Lower organic matter also limits biofilm growth, helping reduce opportunistic pathogens in distribution systems.

The ICRTSD represents the most extensive evaluation of GAC efficacy for treating waters containing elevated TOC and bromide concentrations in the US. The national source water quality and treatment conditions preceding GAC have presumably changed since the data were collected two decades ago, which could influence the characterization herein. Nonetheless, the findings of this paper challenge the presumption that GAC should not be used to treat water with high bromide because of the increased BIF. Rather, this analysis indicates that GAC can be a viable approach to reducing potential risks associated with Br-DBPs in waters containing high bromide. As water utilities are increasingly exploring other types of source waters including higher salinity and reused waters, the high levels of bromide in these waters will pose water quality challenges. While the results of this analysis could inform treatment conditions favorable for Br-THM3 removal by GAC, site-specific differences in pretreatment and TOC composition can cause significant deviations. Bench- or pilot-scale adsorption tests are strongly encouraged for more thorough investigation, especially when multiple treatment objectives are to be met.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1.
GAC influent TOC and bromide concentrations

Figure 2.
Chloroform and Br-THM3 formation in GAC influent

Figure 3.
THM BIF values of GAC influent and effluent at 1- and 2-mg/L TOC breakthrough

Figure 4a.
Chloroform and Br-THM3 formation in GAC effluent 1 vs. Distribution System THM4

Figure 4b.
 Effects of GAC on chloroform and Br-THM3 formation at 1-mg/L TOC breakthrough

Figure 5a.

Chloroform and Br-THM3 formation in GAC effluent 2 vs. Distribution System THM4

Figure 5b.
Effects of GAC on chloroform and Br-THM3 formation at 2-mg/L TOC breakthrough

Figure 6.
Effects of GAC on chlorine demand at 1- and 2-mg/L TOC breakthrough

Table 1.

Types of Disinfectants used by Full-Scale Plants Conducting GAC Studies

Residual Disinfectant ^a	Primary Disinfectant ^a				Total
	Free Chlorine	Chloramines	Chlorine Dioxide	Ozone & Chlorine Dioxide	
Free Chlorine	4	0	3	0	7
Chloramines	5	15	4	1	25
Total					32

^a Reported in the Information Collection Rule Treatment Study Database. GAC – granular activated carbon.

Table 2.Water Quality Summary of 33 GAC Studies ^a

Parameter	Unit	All Plants	Free Chlorine Only Plants ^b	Chloramines Plants ^c
Full-Scale Plant Raw and Finished Water				
Number of plants		32	7	25
Raw water bromide	µg/L	20-1,110 (14655)	20-50 (32/27)	20-1,110 (178/89)
Raw water UV254	l/cm	0.053-0.402 (0.162/0.144)	0.094-0.239 (0.157/0.150)	0.053-0.402 (0.1640.144)
Raw water TOC	mg/L	3.2-9.1 (5.6/5.2)	5.1-7.1 (5.9/6.1)	3.2-9.1 (5.5/5.0)
Raw water TSUVA	L/mg-m	1.9-4.4 (2.9/2.8)	1.5-3.7 (2.7/2.6)	1.5-4.4 (2.9/2.9)
Finished water UV254	l/cm	0.028-0.103 (0.065/0.062)	0.028-0.448 (0.041/0.041)	0.042-0.103 (0.072/0.071)
Finished water TOC	mg/L	2.1-4.8 (3.4/3.4)	2.4-3.4 (2.9/2.8)	2.1-4.8 (3.6/3.6)
Finished water TSUVA	L/mg-m	1.2-2.8 (1.9/1.9)	1.2-1.5 (1.4/1.4)	1.3-2.8 (2.0/2.0)
UV254 reduction	%	1.9-86 (54/59)	49-86 (71/71)	1.9-86 (48/52)
TOC removal	%	2.3-68 (36/37)	44-58 (51.49)	2.3-68 (32/27)
THM4	µg/L	2.6-105 (53/49)	45-105 (71/66)	2.6-102 (48/43)
HAA5	µg/L	3.1-70 (33/29)	22-59 (42/40)	3.1-70 (30/23)
GAC Influent				
Number of studies		33	8	25
Influent bromide	µg/L	20-1,490 (154/62)	20-80 (37/24)	20-1,490 (192/87)
Influent UV254	l/cm	0.024-0.111 (0.064/0.061)	0.040-0.063 (0.052/0.054)	0.024-0.111 (0.068/0.065)
Influent TOC	mg/L	1.8-5.3 (3.5/3.4)	2.4-3.6 (2.9/2.7)	1.8-5.3 (3.6/3.6)
Influent TSUVA	L/mg-m	1.2-2.3 (1.9/1.9)	1.3-2.1 (1.8/1.9)	1.3-2.3 (1.9/1.9)
SDS-CHCl ₃	µg/L	0.4-198 (52/44)	24-152 (58/45)	0.4-198 (50/42)
SDS-BDCM	µg/L	1.5-62 (21/18)	1.7-26 (14/12)	1.5-62 (24/18)
SDS-DBCM	µg/L	0.1-86 (19/12)	0.1-13 (5.3/3.1)	1.8-86 (24/13)
SDS-CHBr ₃	µg/L	0.0-153 (11/0.6)	0.0-1.0 (0.2/0.0)	0.0-153 (15/1.4)
SDS-Br-THM3	µg/L	5.0-200 (52/34)	5.0-37 (19/15)	10-200 (63/37)
SDS-THM4	µg/L	29-234 (104/90)	29-178 (77/67)	33-234 (112/98)
SDS-BIF		0.1-2.8 (0.8/0.5)	0.2-0.6 (0.4/0.4)	0.1-2.8 (0.9/0.8)
Chlorine demand	mg/L as Cl ₂	1.4-5.6 (3.0/2.8)	1.4-4.6 (2.7/2.6)	1.9-5.6 (3.1/2.8)
GAC Effluent at 1-mg/L TOC Breakthrough				
UV254	l/cm	0.004-0.018 (0.011/0.012)	0.007-0.018 (0.011/0.011)	0.004-0.018 (0.011/0.012)
TSUVA	L/mg-m	0.4-1.8 (1.1/1.2)	0.7-1.8 (1.1/1.1)	0.4-1.8 (1.1/1.17)
UV254 reduction	%	54-94 (81/82)	67-88 (77/79)	54-94 (82/83)
TOC removal	%	45-81 (70/71)	58-73 (65/63)	45-81 (71/72)
SDS-CHCl ₃	µg/L	0.0-27 (5.6/4.4)	1.4-23 (8.2/6.0)	0.0-27 (4.8/3.8)
SDS-BDCM	µg/L	0.0-23 (6.4/6.3)	2.4-11 (5.8/6.5)	0.0-23 (6.5/6.3)
SDS-DBCM	µg/L	0.3-18 (7.4/6.0)	1.2-11 (5.4/4.8)	0.3-18 (8.0/7.4)

Parameter	Unit	All Plants	Free Chlorine Only Plants ^b	Chloramines Plants ^c
SDS-CHBr ₃	µg/L	0.0-29 (7.3/2.8)	0.0-4.5 (1.2/0.4)	0.0-29 (9.3/5.8)
SDS-Br-THM3	µg/L	1.7-50 (21/18)	3.6-20 (13/12)	1.7-50 (24/22)
SDS-THM4	µg/L	1.7-51 (27/25)	8.1-40 (21/22)	1.7-51 (29/27)
SDS-BIF		0.4-3.0 (1.5/1.4)	0.6-1.7 (1.0/0.9)	0.4-3.0 (1.7/1.5)
SDS-Br-THM3 removal	%	0-94 (42/41)	6-46 (30/31)	0-94 (47/45)
SDS-THM4 removal	%	42-96 (72/73)	59-78 (72/73)	42-96 (72/73)
Chlorine demand	mg/L as Cl ₂	0.5-2.0 (1.1/1.1)	0.7-1.5 (1.1/1.1)	0.5-2.0 (1.1/1.1)
Chlorine demand reduction	%	27-82 (63/64)	50-69 (57/55)	27-82 (65/69)
GAC Effluent at 2-mg/L TOC Breakthrough				
UV254	1/cm	0.015-0.039 (0.026/0.026)	0.018-0.031 (0.025/0.027)	0.015-0.039 (0.026/0.026)
TSUVA	L/mg-m	0.8-2.0 (1.3/1.3)	0.9-1.6 (1.3/1.4)	0.8-2.0 (1.3/1.3)
UV254 reduction	%	27-77 (57/59)	43-59 (51/51)	27-77 (59/62)
TOC removal	%	17-62 (41/41)	17-45 (31/29)	20-62 (44/45)
SDS-CHCl ₃	µg/L	0.0-75 (17/14)	8.4-60 (23/16)	0.0-75 (15/11)
SDS-BDCM	µg/L	0.1-38 (13/12)	3.1-19 (12/12)	0.1-38 (13/12)
SDS-DBCM	µg/L	1.2-48 (14/12)	1.2-17 (8.4/6.7)	3.1-48 (16/13)
SDS-CHBr ₃	µg/L	0.0-73 (12/3.5)	0.0-4.3 (1.2/0.1)	0.0-73 (15/4.9)
SDS-Br-THM3	µg/L	5.0-119 (39/30)	5.0-39 (21/18)	9.7-119 (44/34)
SDS-THM4	µg/L	19-121 (55/48)	19-85 (44/41)	22-121 (59/51)
SDS-BIF		0.3-2.9 (1.2/1.0)	0.3-1.2 (0.7/0.8)	0.3-2.9 (1.4/1.3)
SDS-Br-THM3 removal	%	-41-69 (11/12)	-41-9 (-12-1)	-6-69 (20/14)
SDS-THM4 removal	%	23-71 (46/47)	23-53 (40/40)	26-71 (48/48)
Chlorine demand	mg/L as Cl ₂	0.8-2.8 (1.6/1.6)	0.9-2.3 (1.5/1.4)	0.8-2.8 (1.6/1.6)
Chlorine demand reduction	%	6-67 (46/47)	33-56 (44/44)	6-67 (46/49)

^a Range (mean/median) values are presented in the table; computed based on plant annual averages and averages of RSSCT or pilot test runs for each study.

^b Plant used free chlorine for primary and residual disinfection.

^c Plant used chloramines for residual disinfection.

GAC – granular activated carbon, UV254 – ultraviolet absorbance at 254 nm, TOC – total organic carbon, TSUVA – specific ultraviolet absorbance at 254 nm (normalized for TOC), SDS – simulated distribution system, CHCl₃ – chloroform, BDCM – bromodichloromethane, DBCM – dibromochloromethane, CHBr₃ – bromoform, Br-THM3 – sum of three brominated trihalomethanes, THM4 – sum of four trihalomethanes, HAA5 – sum of five haloacetic acids, BIF – bromine incorporation factor.