Removal of algal taste and odour compounds by granular and biological activated carbon in full-scale water treatment plants

Aisha Faruqi, Milann Henderson, Rita K. Henderson, Richard Stuetz, Brendan Gladman, Bridget McDowall and Arash Zamyadi

ABSTRACT

The occurrence and severity of cyanobacterial and algal blooms in water supplies has been increasing due to the effects of eutrophication and climate change, resulting in more frequent taste and odour (T&O) events. Conventional treatment processes have been found to be inefficient in removing the two most commonly detected algal T&O compounds, geosmin and 2-methylisoborneol (MIB), though granular activated carbon (GAC) and biological activated carbon (BAC) contactors have achieved high T&O removal rates. Literature on the performance of GAC and BAC for T&O removal in full-scale treatment plants, however, is limited. This review collates and assesses pilot-scale and full-scale studies which focus on removal of geosmin and MIB, with the aim of understanding the factors which influence T&O removal and determining knowledge gaps in the use of GAC and BAC. Age and empty bed contact time (EBCT) were found to have a significant impact on GAC performance, with removal efficiency decreasing with increased age and increasing with longer EBCTs. BAC contactors have achieved higher removal rates than non-biologically active GAC contactors and were not impacted by age, EBCT and/or carbon type. From these observations, implementation of BAC for T&O removal would be favourable; however, further investigations are required to understand full-scale performance of BAC and service life modelling.

Key words | 2-methylisoborneol (MIB), biological activated carbon (BAC), cyanobacteria, geosmin, granular activated carbon (GAC), taste and odour

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INTRODUCTION

In recent years, incidences of cyanobacterial/algal bloom detection have increased worldwide (Zamyadi 2014). Cyanobacteria predominantly cause concern as producers of toxic or taste and odour (T&O) compounds, and their increasing presence and impact have been well-documented (Ho & Newcombe 2010; Zamyadi 2014; Park *et al.* 2015). Cyanotoxins threaten wildlife, domestic animals and humans, most commonly through consumption of contaminated water (Svrcek & Smith 2004; Zamyadi 2014). In water sources, 2-methylisoborneol (MIB – $C_{11}H_{20}O$) and geosmin ($C_{12}H_{22}O$) (Figure 1) are two commonly detected cyanobacterial T&O compounds (Zamyadi *et al.* 2015a, 2015b). While several studies focus on the removal of cyanotoxins, literature on T&O removal is limited.

Geosmin and MIB (Figure 1) contribute musty, earthy tastes and odours to water. Undesirable taste and odour characteristics of drinking water can cause significant concern to the public, and are a major source of complaints made to water utilities by customers each year (Park *et al.* 2015; Zamyadi *et al.* 2015a, 2015b). Although not of health

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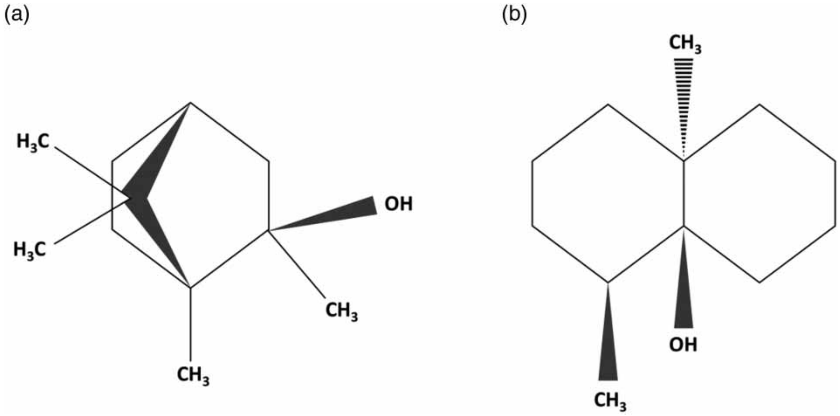

Figure 1 | Chemical structure of (a) 2-methylisoborneol (MIB) and (b) geosmin.

concern, unpleasant taste or odour of treated water may be associated with poor quality by the customer and lead to mistrust of the water providers (Ho & Newcombe 2010). MIB and geosmin can be detected as an odour and/or a taste by humans in very low concentrations even below 10 ng/L (Young *et al.* 1996; Omur-Ozbek *et al.* 2007).

Several factors influence the growth of cyanobacteria and these must be considered when predicting and managing levels of T&O in water treatment plants. Eutrophication and climate change, specifically warming and altered rainfall patterns, have been identified as two major factors which promote the growth of cyanobacteria, and subsequently cause cyanobacterial blooms in water bodies (Paerl & Paul 2012; Sinha *et al.* 2012). However, it has also been suggested that at least a portion of the perceived increase in cyanobacterial growth may be attributed to increased monitoring (Sinha *et al.* 2012). Bloom events are characterised by the accumulation of cyanobacterial cells, forming dense scums at the water's surface, and are enhanced in stagnant waters with long residence times and at a neutral to alkaline pH (6–9) (Svrcek & Smith 2004). Cell lysis results in an increased release of toxic and T&O compounds from the cell during and after a bloom event (Svrcek & Smith 2004; Zamyadi 2014).

Increased intensity and altered frequency of rain events deliver nutrients to surface water via run-off, contributing to growth of cyanobacterial blooms. This can be exacerbated once flood events subside, and the residence time of surface waters increase; a problem that is particularly evident during drought periods. Increased severity of droughts and subsequent pressure on freshwater resources resulting in salinisation may also favour cyanobacterial dominance of water sources through exclusion of phytoplankton species with low salinity tolerances (Paerl & Paul 2012). As such, climate change and its effects will likely continue to contribute to the increased prevalence of cyanobacteria and their harmful metabolites for the foreseeable future.

Seasonal fluctuations in cyanobacterial presence in water bodies and subsequently in MIB and geosmin concentrations have been observed. In a study of lakes in semi-arid regions of Arizona, Westerhoff *et al.* (2005) observed and explained these trends. Throughout spring to late summer, MIB and geosmin concentrations were highest, with stratification of the water column causing the highest concentration of T&O compounds to occur in the epilimnion (which is the upper layer of water in a stratified lake). Destratification in autumn resulted in dilution of the epilimnion and increased MIB and geosmin presence in the water profile. Implementation of cost-effective treatment barriers to safeguard the drinking water against these seasonal variations is one of the major challenges faced by water utilities (Westerhoff *et al.* 2005).

T&O compounds are difficult to remove using conventional treatment processes such as clarification, filtration and oxidation using chlorine (Lalezary *et al.* 1986; Zamyadi *et al.* 2015a). As such, removal through adsorption, which is much more efficient (Ho & Newcombe 2010; Zamyadi *et al.* 2015a), is becoming increasingly popular within the water treatment industry. While powdered activated carbon (PAC) can be used for adsorption during earlier stages of the treatment train, granular activated carbon (GAC) filter media has been found to be the most effective method of adsorption, as well as biodegradation which occurs in biological activated carbon (BAC) (Ahn *et al.* 2007).

While several studies are available on removal of T&O compounds using adsorption on both virgin GAC and PAC, information about the removal capacity of BAC is limited. Additionally, gaps exist in the literature regarding the removal efficiency of both GAC and BAC as they age. The adsorption capacity of activated carbon tends to decrease as it ages, resulting in an increased risk of breakthrough of T&O compounds. This calls for a method to understand activated carbon performance over time so that water utilities can accurately predict when renewal of activated carbon contactors is needed. There is currently also very limited data on the performance of GAC and BAC in full-scale treatment plants, with most studies employing bench-scale and pilot-scale tests. The aim of this review is to determine the knowledge gaps in assessing the capacity of granular and BAC to remove T&O compounds during water treatment. To the best of the authors' knowledge, this is the first review of its kind focusing on the fate of these compounds and performance of BAC in full-scale water treatment plants.

DATA COLLECTION: METHODOLOGICAL APPROACH

A literature review was carried out starting with collection of available data on the use of GAC and BAC for T&O removal, with the focus being on pilot- and full-scale studies and/or use of natural water/bloom samples. Of the studies reviewed, 48% of them analysed bench-scale laboratory experiments, 44% used detailed pilot-scale experiments and only 8% of analysed results were obtained in a full-scale water treatment plant (WTP). This is attributed to the increased cost, risk and time required to build and run a full-scale experiment. 16% of these studies focused specifically on biodegradation in laboratory-scale sand filter columns, batch biofilm degradation experiments, and analysing T&O compound trends in reservoirs.

Data from bench-, pilot- and full-scale studies have been sourced from the USA (Illinois, Florida, Pennsylvania, Colorado, Minnesota), Canada (Toronto, Lake Ontario), Sweden (Gothenburg), Australia (Murray River, Morgan, Adelaide, Armidale), China (Kinmen County), and South Korea (Gwanju) (Gillogly *et al.* 1999; Nerenberg *et al.* 2000; Elhadi 2004; MacKenzie *et al.* 2005; Ndiongue *et al.* 2006; Ho *et al.* 2007; Persson *et al.* 2007; McDowall 2008; Drikas *et al.* 2009; Hoefel *et al.* 2009; McDowall *et al.* 2009; Thompson & Brooks 2009; Ho & Newcombe 2010; Scharf *et al.* 2010; Yang *et al.* 2010; Jakubowski *et al.* 2011; Smith *et al.* 2012; Zhang 2012; Summers *et al.* 2013; Greenwald *et al.* 2015; Li 2015; Park *et al.* 2015; Zamyadi *et al.* 2015a). The geographical locations of pilot- and full-scale studies assessed in this review are presented in Figure 2.

Data from the reviewed studies were collated to determine the impact of GAC and BAC age and empty bed contact time (EBCT) on removal efficiency of MIB and geosmin. Removal efficiencies achieved using a similar EBCT of 4–5 min were compared to show their variation with GAC age. Studies using higher EBCTs were not included to ensure that variations in removal efficiency were due to age and not differences in EBCT. To assess the impact of EBCT on removal efficiency, the different GAC and BAC materials were grouped by age, and their removal efficiencies were compared by EBCT. MIB and geosmin removal by GAC was primarily through adsorption whereas removal by BAC was through both adsorption and biodegradation.

FACTORS INFLUENCING GAC AND BAC REMOVAL EFFICIENCY AND CAPACITY

This review focuses on results obtained from pilot- and full-scale studies to assess the factors which influence GAC performance. Pilot-scale testing has been found to be more accurate than bench-scale methods which can sometimes overestimate GAC adsorption capacity (Li 2015; Zamyadi *et al.* 2015a, 2015b). Pilot-scale testing utilises GAC particles that are the same size as those used in full-scale columns, whereas bench-scale tests such as rapid small scale column tests involve crushing GAC particles to a smaller size, influencing rate of adsorption and/or characteristics of the biofilm (Corwin & Summers 2010; Kennedy *et al.* 2015). Ho & Newcombe (2010) used large scale columns (Diameter 25 cm and over) during a pilot-scale study which allows the application of EBCT and GAC volume similar to full-scale contactors. This set-up provided the

Figure 2 | Geographical locations of the pilot- and full-scale studies presented in this literature review.

opportunity to accurately simulate and model full-scale GAC contactors.

It was found that a significant number of factors influence the removal efficiency and capacity of GAC for MIB and geosmin removal. On the whole, rates of geosmin removal exceeded rates of MIB removal for all waters and GAC types (Elhadi 2004; Li 2015; Zamyadi *et al.* 2015a, 2015b). The effects of GAC characteristics, water characteristics, biological activity, EBCT, regeneration and experimental design on MIB and geosmin removal have been studied in the literature, with varying degrees of success. These are explored and assessed below.

Activated carbon characteristics

Coal-based GAC is by far the most studied GAC type, with limited studies considering each of wood-based and coconut-based GACs. Several studies observing the efficiency of biological activity for the removal of T&O compounds in sand and anthracite filters were also reviewed (Elhadi 2004; Ho *et al.* 2007; McDowall 2008; McDowall *et al.* 2009). While most GAC manufacturers provide a range of qualitative and quantitative parameters that classify their products, it is often unclear exactly what these characteristics indicate in terms of GAC and particularly BAC application and performance.

Trace capacity number (TCN), Brunauer, Emmett and Teller (BET) surface area, and Iodine number are common characteristics provided by GAC suppliers as characterisation of different GACs. Smith *et al.* (2012) showed that TCN should not be relied upon as a predictor of GAC performance, being only very weakly correlated to MIB breakthrough ($R^2 < 0.05$). On the other hand, MIB isotherm tests, BET surface area and Iodine number did exhibit strong positive correlations ($R^2 > 0.9$), with the exception of one outlier – a lignite coal charcoal in a test otherwise composed of bituminous coals. This may be explained by the higher transport pore volume (mL/g) of the lignite coal, being two times greater than the next highest transport pore volume. There were no strong correlations between MIB loading at 50,000 bed volumes and any of these parameters. These results, however, should be considered with caution, given the low sample size of only five GACs in the study by Smith *et al.* (2012).

Additionally, work by Greenwald *et al.* (2015) highlighted the fact that different GAC source materials (e.g. coconut, bituminous coals) yield different critical pore volume ranges, and these may provide crucial information about the lifespan, suitability and efficiency of a GAC product. Activated carbons with hydrophobic surfaces, for example coconut based GAC, adsorb MIB better than non-hydrophobic carbons such as wood based GAC (Pendleton *et al.* 1997). Furthermore, Mackenzie *et al.* (2005) concluded that surface chemistry of carbon is more important for reactivated carbons. Li's (2015) observation of carbon surface under electronic microscope demonstrated that virgin GAC has more disordered surfaces, with a clear porous structure, while the surface of aged GAC is more flat indicating blocked adsorption sites.

Water characteristics

It has been determined in several studies that removal of MIB and geosmin by aged GAC is determined not by MIB and geosmin concentrations but rather by natural organic matter (NOM) loading on the GAC surface or presence in the water influent. Adsorption of both geosmin and MIB onto GAC is negatively influenced by the presence and quantity of organic matter (Gillogly *et al.* 1999; Summers *et al.* 2013; Greenwald *et al.* 2015; Park *et al.* 2015), the adsorption of which in turn is influenced by water temperature (Persson *et al.* 2007).

Ho & Newcombe (2010) identified pore blockage, and reduction in available high-energy adsorption sites as the GAC ages, as two mechanisms by which organic matter decreases removal efficiency of T&O compounds by GAC. Li (2015) also hypothesised that NOM fouling created a film on the GAC surface, resulting in the blockage of active sites.

It may be advantageous to limit organic matter in the influent to GAC columns to extend life of the bed for removal of T&O compounds by adsorption. However, the availability of biodegradable organic matter (BOM) may be crucial to the establishment and support of an active biofilm (Elhadi 2004). This should be carefully considered alongside biological activity as a method to enhance GAC removal of MIB and geosmin. However, further studies are required to expand on the requirement for BOM in developing biomass and BAC performance.

In the same study by Elhadi (2004), temperature was shown to have a substantially larger effect on MIB and geosmin removal than media type or influent MIB and geosmin concentration. Removal by biologically active GAC was seen to be reduced by up to 40% with a decrease in temperature from 20 °C to 8 °C (Elhadi 2004). Biological activity may not be a reliable method of removal of MIB and geosmin in cold seasons or climates for this reason, although seasonality of MIB and geosmin concentration may result in high concentrations of these T&O compounds occurring in warm periods.

It is commonly reported in the literature that geosmin is more readily removed than MIB, whether through adsorption or biodegradation (Elhadi 2004; Summers *et al.* 2013). This knowledge may be incorporated into experimental design where it is desired to remove similar concentrations of MIB and geosmin from influent water. If a given concentration of MIB removal is achieved, it should be expected that the same, or better, removal of geosmin would be achieved. An understanding of the higher difficulty of removal of MIB should also be incorporated into plant design, with GAC adsorbers designed to at least control maximum expected influents of MIB.

Impact of biological activity and biodegradation process

GAC filters that are biologically active have been found to achieve higher removal rates of MIB and geosmin in comparison with GAC contactors in which adsorption is the primary removal method (Figure 3). As virgin porous carbon (Figure 3(a)) is used for water treatment, NOM and microbial community fill in the pores within carbon until all the adsorption sites are completely exhausted (Figure 3(b)). In case an active microbial community forms the biofilm a GAC contactor would turn into a BAC system. Activity of a biofilm follows two simultaneous phases: substrate diffusion and biological reaction. Biofilm diffusion profiles are parabolic, as electron donors and acceptors diffuse from bulk liquid into the biofilm, microbial cells within the biofilm metabolise them (Zhu *et al.* 2010). BAC data presented in this review is of GAC columns which the authors have deemed as biologically active. Adenosine triphosphate analysis, bacterial enumeration (for example, by flow cytometry) and scanning electron

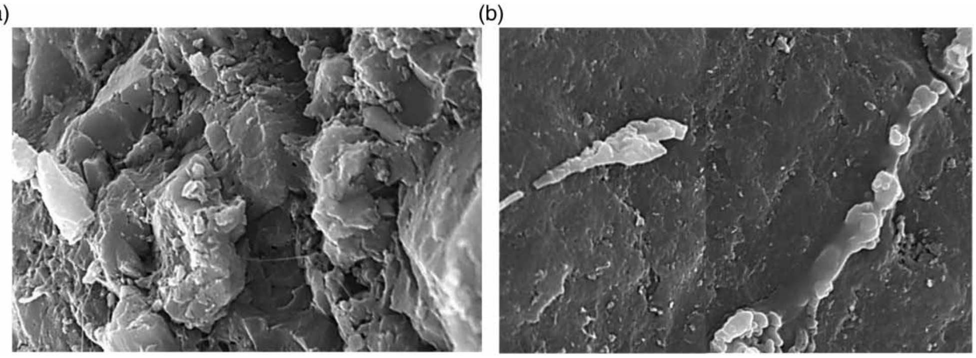

Figure 3 | GAC surface observation using scanning electron microscopy (SEM – ×5,000): (a) new carbon (less than 5,000 bed volume) and (b) aged carbon (over 90,000 bed volume) (Images' copyright: A. Zamyadi).

microscopy (SEM) are commonly used analyses for quantifying and describing the presence, composition and activity of biofilms and their constituents (McDowall 2008; Zamyadi *et al.* 2015a, 2015b). While the precision and accuracy of these methods is not the focus of this literature review, Zhu *et al.* (2010) suggested that combination of genotypic and phenotypic analysis could help discovering biofilm phylogenetic traits and metabolic characteristics. Ho *et al.* (2007) demonstrated the effectiveness of biological degradation for complete removal of MIB and geosmin from water using sand filters (media with none or very low adsorption capacity), where biodegradation was the primary removal mechanism. This study found that removal of MIB and geosmin was not dependent on their initial concentrations (50 and 100 ng/L). Biodegradation rate is, however, dependant on initial microbial concentration of the biofilm on the sand filter (Ho *et al.* 2007), and temperature (Hoefel *et al.* 2009), thus highlighting importance of understanding and modelling the biological activity.

Experiments carried out on non-adsorptive filter material (for example, sand) further exemplify the potential of biological activity for increased removal of MIB and geosmin. McDowall (2008) assessed a full-scale water treatment plant in South Australia where an operational change led to chlorinated water being used in backwash of 26-year-old sand filters that had previously been reported to remove MIB and geosmin. Subsequent to the chlorinated backwash, breakthrough of MIB and geosmin was observed in the effluent water. Reversion to non-chlorinated backwash water saw an improvement in MIB and geosmin removal and effluent concentrations of both taste and odour compounds returned to below 5 ng/L.

Laboratory-scale bench studies on sand filters further showed that age of the biofilm is an important factor in its resistance to changing operation conditions and influent concentrations. Newly established biofilms are susceptible to fluctuations in removal efficiency following changes in EBCT and periods of low influent T&O compound concentration, and require a longer acclimation period before a steady-state removal is reached when compared with aged biofilms (McDowall 2008). Although biologically active sand/anthracite filters were shown to remove MIB and geosmin in this study, experiments carried out by Elhadi (2004) showed that biofilms on GAC require a shorter acclimation period, are more robust than biofilms on anthracite or sand, and that a greater biomass may be supported on GAC than on anthracite. However, biofilms on anthracite are less susceptible to change in temperature than biofilms on GAC (Elhadi 2004).

Pseudomonas sp., *Alphaproteobacterium* sp., *Sphingomonas* sp. and *Acidobacteriaceae* sp. have been identified as microorganisms to be involved significantly in cooperative biodegradation of MIB and geosmin (Ho *et al.* 2007) and Geo24, a gram-negative *Sphingopyxis* sp., has been found to be capable of sole degradation of geosmin (Hoefel *et al.* 2009), though this list is by no means exhaustive. Removal of MIB and geosmin may be increased by up to 40%, through seeding of columns with combinations of specific MIB- or geosmin-degrading bacteria as shown by McDowall *et al.* (2009). Seeding of columns is most effective when a biofilm, either active or inactive, is present, and may also decrease acclimation periods and prevent lag after a period of low T&O compound concentration in the influent water.

Zamyadi et al. (2015a) found that BAC can achieve up to 40% greater removal of MIB and 5% greater removal of geosmin than sterile GAC in bench-scale tests. Similarly, Ho & Newcombe (2010) measured up to 30% reduction in removal of MIB when chlorine was measured in the influent water. Pilot tests have indicated that two-year-old BAC retains the capacity to remove 60% of MIB and 80% of geosmin (Zamyadi et al. 2015a). Gillogly et al. (1999) stipulate that, without biological activity, an EBCT of less than 10 min will be inadequate for MIB and geosmin removal. Further pilot-scale testing has found BAC to be effective at removing sufficient amounts of geosmin (Strait 2015). While influent concentrations of geosmin were not steady, GAC types from different manufacturers used for biofiltration by Strait (2015) were able to reduce the amount of geosmin present in the water to below the threshold limit (4 ng/L) for the most part of the study. Measurement of biofilm activity is a limiting factor while studying biological degradation of the cyanobacterial metabolites (McDowall 2008; Zamyadi et al. 2015b). Formation and maintenance of an active biofilm is a challenge during the operation of biologically active water treatment processes. The development of rapid methods to monitor the biofilm formation, composition, and level of activity would provide the utilities with essential operational information. Treatment methods used prior to filtration through BAC must also be considered as pre-chlorination limits biological film growth and oxidises the GAC surface, reducing the ultimate capacity of the GAC for MIB and geosmin removal (Gillogly et al. 1999; Ho & Newcombe 2010).

Biological removal of T&O compounds, in conjunction with adsorption, may be desirable as a low cost, low maintenance option that reduces chemical sourcing or equipment maintenance requirements. When used in conjunction with adsorption, it can increase removal of T&O compounds to below detection limits, which is particularly useful in aged GACs. However, biodegradation may be difficult to predict, is dependent on an array of water, site, and biological community characteristics, and can require significant establishment time for growth of an effective biofilm (McDowall et al. 2009).

Impact of pre-oxidation focusing on biofilm enhancement

Certain pre-oxidation practices are known to enhance removal of MIB and geosmin during water treatment (Yang et al. 2010; Jakubowski et al. 2011; Zamyadi et al. 2015b). Agents such as ozone (Park et al. 2015) or combined processes like ultraviolet (UV)/H_2O_2 (Zamyadi et al. 2015b) may be used to achieve oxidation in water treatment plants prior to GAC. Ozonation can destroy a significant percentage of MIB and geosmin compounds present in water (Liang et al. 2007). As well as being able to partially remove T&O compounds, pre-ozonation can positively impact biofiltration by assisting with growth of a biofilm on GAC through oxidation of NOM present in the water (De Waters & DiGiano 1990).

Figure 4 shows the removal efficiencies of ozone/BAC systems and the contribution of biofiltration to removal in these systems. All studies shown in Figure 4 achieved geosmin and MIB removals of over 90% with combined ozonation and biofiltration. Ozonation improved T&O

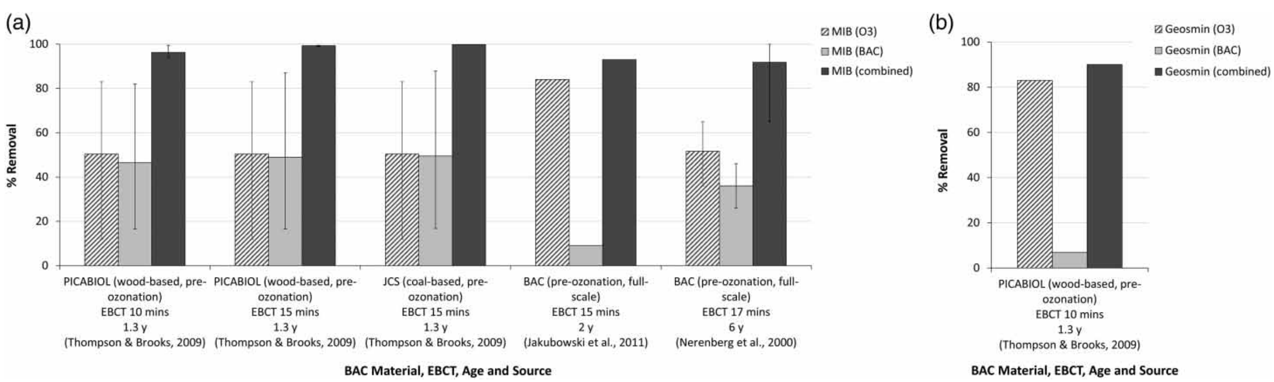

Figure 4 | Contribution of both ozonation and bio-filtration to removal of (a) MIB and (b) geosmin in O_3/BAC systems.

compounds removal by (a) direct oxidation of these compounds and (b) enhanced biological activity. In a full-scale study conducted by Jakubowski *et al.* (2011), ozonation was found to be responsible for removal of 83% of geosmin and MIB in settled water (Figure 4). Yang *et al.* (2010) also assessed a pilot-scale O_3/BAC system, achieving 96.3% removal of geosmin and over 99% removal of MIB (Figure 4). They found that BAC treating pre-ozonated water achieved 3% greater MIB removal and 15% greater geosmin removal compared to comparable GAC columns without pre-ozonation. A pilot-scale study of ozonation followed by BAC filters by Edwards-Brandt *et al.* (2007) also found ozonation to be sufficient for removal of geosmin and, to a lesser extent (10%), MIB.

BAC contactors consisting of selective biofilm microbial communities adapted to cyanobacterial/algal T&O compounds have been shown to be sufficient barriers for removal of these compounds to below detection limits. For example, studies utilising BAC without pre-ozonation, presented further in this review, have achieved removals of greater than 95%, suggesting that pre-ozonation may not be required if T&O removal is the sole purpose of implementing BAC filters (Zhu *et al.* 2010; Park *et al.* 2015). Park *et al.* (2015) further showed that when influent water geosmin levels are below 100 ng/L, treatment with O_3, H_2O_2 or a combination of both is unnecessary, as BAC is capable of removing this concentration to below taste and odour thresholds effectively. MIB removal, however, is likely to be lower, and as such, ozonation may be beneficial even at lower influent concentrations. Implementation of ozonation treatment may not be viable or justified unless influent MIB and geosmin concentrations are consistently or frequently above 100 ng/L. In addition, efficient application of methods to enhance microbial adhesion for removal of target T&O compounds, such as biofilm thickness control and microbial community manipulation (including backwash frequency and carbon surface alterations), are still significant operational challenges (Zhu *et al.* 2010; Zamyadi *et al.* 2015a).

Activated carbon age

The age of GAC used for taste and odour removal has been found to impact its removal efficiency, with an increase in age resulting in decreased removal of MIB and geosmin. Figures 5 and 6 show this trend among GAC columns utilising similar empty bed contact times of between 4 and 5 min for removal of MIB and geosmin, respectively. Both figures show a variation in capacity of GAC for removal of MIB and geosmin with respect to age and type of GAC, as well as type of pre-treatment used prior to GAC filtration. Adsorption capacity decreases with time, with a virgin GAC achieving 92% MIB removal while a 4-year-old GAC achieved only 15% removal of MIB (Figure 5). It should be noted that the data presented is sourced from several studies, each with unique source water characteristics. This can significantly impact results, as although influent MIB concentration does not have a significant impact on percentage removal, dissolved organic matter in source water does decrease GAC removal capacity (Summers *et al.* 2013).

Figure 5 shows a significant difference in MIB removal capacity of two different 1-year-old GACs. A 1-year-old catalytic (chemical alteration of carbon surface characteristics) GAC tested by Zamyadi *et al.* (2015a) achieved a removal

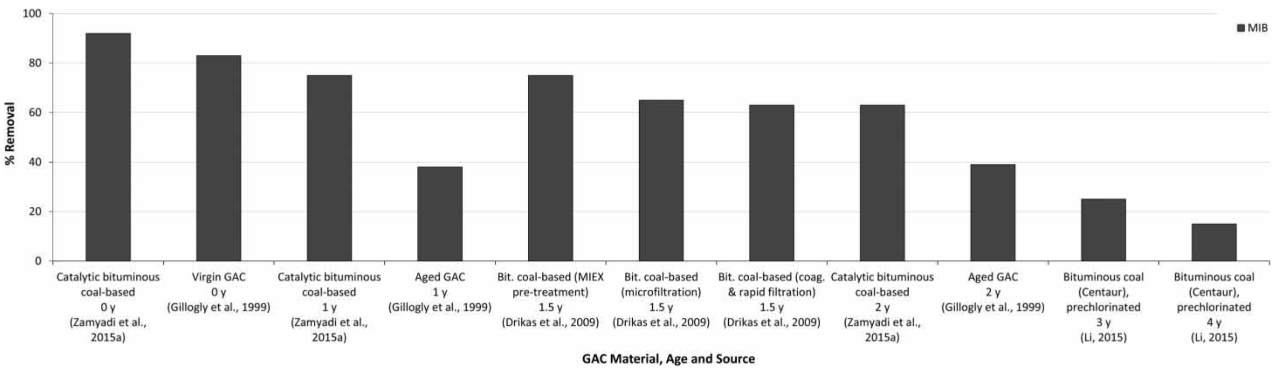

Figure 5 | Impact of age on MIB removal efficiency of GAC with an EBCT of 4–5 min.

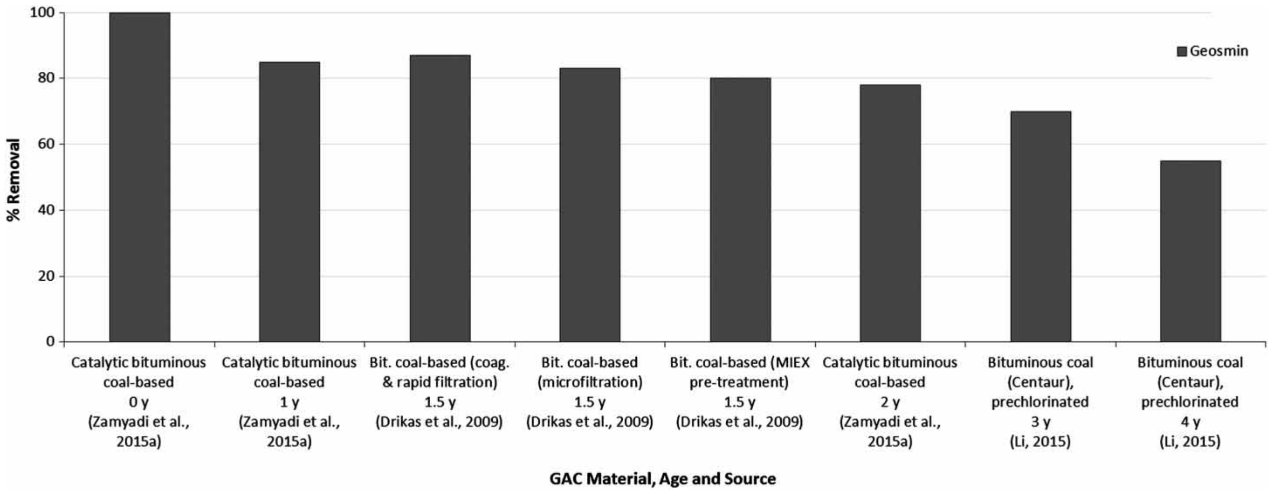

Figure 6 | Impact of age on geosmin removal efficiency of GAC with an EBCT of 4–5 min.

efficiency of 75% while a non-catalytic GAC of the same age, tested by Gillogly *et al.* (1999) achieved a much lower removal efficiency of 38%. The variance could be due to a difference in and dissolved organic carbon (DOC) adsorption by each GAC. While the rate of NOM adsorption was not measured by Gillogly *et al.* (1999), additional experimentation within the same study found a decrease in MIB removal efficiency caused by increased NOM loading (Gillogly *et al.* 1999). As well as the comparatively low removal efficiency being caused by a higher NOM loading, the authors noted that chlorine may also have had an impact on the GAC's removal efficiency as the plant used did utilise pre-chlorination.

The similar removal efficiencies of MIB attained by a 1-year-old and 1.5-year-old GAC (Figure 5) could be the result of low NOM loading (Drikas *et al.* 2009). An older GAC would be expected to have a lower T&O removal efficiency due to a decrease in available adsorption sites. However, as the water prior to 1.5-year bituminous coal was pre-treated using MIEX resin, DOCs were removed from the water, resulting in fewer adsorption sites being occupied by NOM. The use of MIEX in the pre-treatment phase and resulting lower DOC concentration also explains the GAC's higher removal efficiency compared to the two other 1.5-year-old GACs (without pretreatment) from the same study (Drikas *et al.* 2009). Figure 6 also shows the same trend of removal capacity decreasing with age, though with geosmin removals consistently remaining much higher than the MIB removal efficiencies (Figure 5) due to its chemical properties.

Few studies on BAC removal efficiency are available; however, two studies which utilised EBCTs of 4–5 min achieved higher MIB and geosmin removals than non-biologically active carbon of a similar age. Three 1.5-year-old BACs examined by Drikas *et al.* (2009), with different pre-treatments, achieved over 99% removal of both MIB and geosmin. Notably, a 2-year-old BAC examined by Scharf *et al.* (2010) with an EBCT of 5.2 min achieved a lower geosmin removal efficiency of 90%, despite being older. This could be due to there being a lower biomass on the 2-year-old BAC as biological activity was only assumed and not confirmed. This BAC did still, however, achieve higher removal efficiency than a 2-year-old GAC which achieved only 78% geosmin removal (Figure 6).

EBCT and bed depth

EBCT is a function of the GAC filter depth and loading rate. This parameter has a significant effect on the performance of GAC adsorbers, with Schideman even classifying it as the most important design criteria (Schideman *et al.* 2012).

Drikas *et al.* (2009) found that GAC had the capacity to remove MIB and geosmin to below detection limits in pilot scale columns at 20 min EBCT for at least 10 months. Decreasing the EBCT to 5 min after 10 months resulted in decreases of up to 50% in the removal capacity of the GAC; a further 8 months of filtration (at the reduced EBCT) saw BAC removal reach 100%, whilst sterile GAC reached only 75% and 80% removal for MIB and geosmin, respectively (at the same EBCT). For 4-year-old GAC, Li (2015) showed 45% greater removal of geosmin for an EBCT of 2 min longer, and a GAC bed twice as deep.

Zamyadi *et al.* (2015a) also found a relationship between EBCT, bed depth, and removal. Measuring MIB and geosmin concentrations at sampling ports along the length of 128 cm long GAC pilot-scale columns, removal was found to increase as bed depth increased. Geosmin removals of 10–20%, 40–60%, and ~80% at bed depths of 25 cm, 100 cm and 128 cm, respectively, were achieved. MIB removals of 10–40%, 40–60% and ~65% were achieved using bed depths of 25 cm, 100 cm and 128 cm, respectively. Notably, the MIB removal percentage using virgin GAC with 75 cm of depth followed a similar range to the aged GAC. An EBCT of 4.1 min and bed depth of 25–50 cm of virgin GAC was sufficient to decrease geosmin concentration to below detection limit in the column effluent. The degree of removal was comparable with that reported by Ndiongue *et al.* (2006), with 61–75% and 24–47% geosmin removal using 25 cm of virgin and used (about 3 years) bituminous GAC, respectively, at 2.8 min EBCT. Furthermore, Ndiongue *et al.* (2006) demonstrated that an increase in GAC depth to 95 cm, and an increase of EBCT from 2.8 min to maximum 7.5 min, significantly increased geosmin removal efficiency of used GAC by around 35%.

Figure 7(a) shows an increase in GAC removal efficiency as EBCT increases. Six-month-old GAC columns with EBCTs of 15 min and greater have achieved removals above 80% while a virgin GAC with an EBCT of 2.6 min achieved less than 35% removal. Despite having a much longer EBCT, a 6-month-old coal-based GAC tested by Park *et al.* (2015) shown in Figure 7(a) achieved a lower geosmin and MIB removal than a virgin coal-based GAC tested by Zamyadi *et al.* (2015a). It can be hypothesised that this lower removal efficiency is due to a difference in surface characteristics as the virgin GAC with a much shorter EBCT was catalytic while the other was not. Furthermore, despite having a longer EBCT, GACs tested by Gillogly *et al.* (1999) achieved lower MIB removal efficiency than expected (Figure 7(a)–7(c)) due to a higher NOM loading on the GAC.

Figure 7(a) and 7(b) also show the impact of operational practices and parameters including pre-treatment on T&O compounds removal efficiency. For example, three coal-based GACs tested by Park *et al.* (2015) achieved different removal efficiencies despite having the same age and EBCT due to pre-oxidation practices (Figure 7(a)).

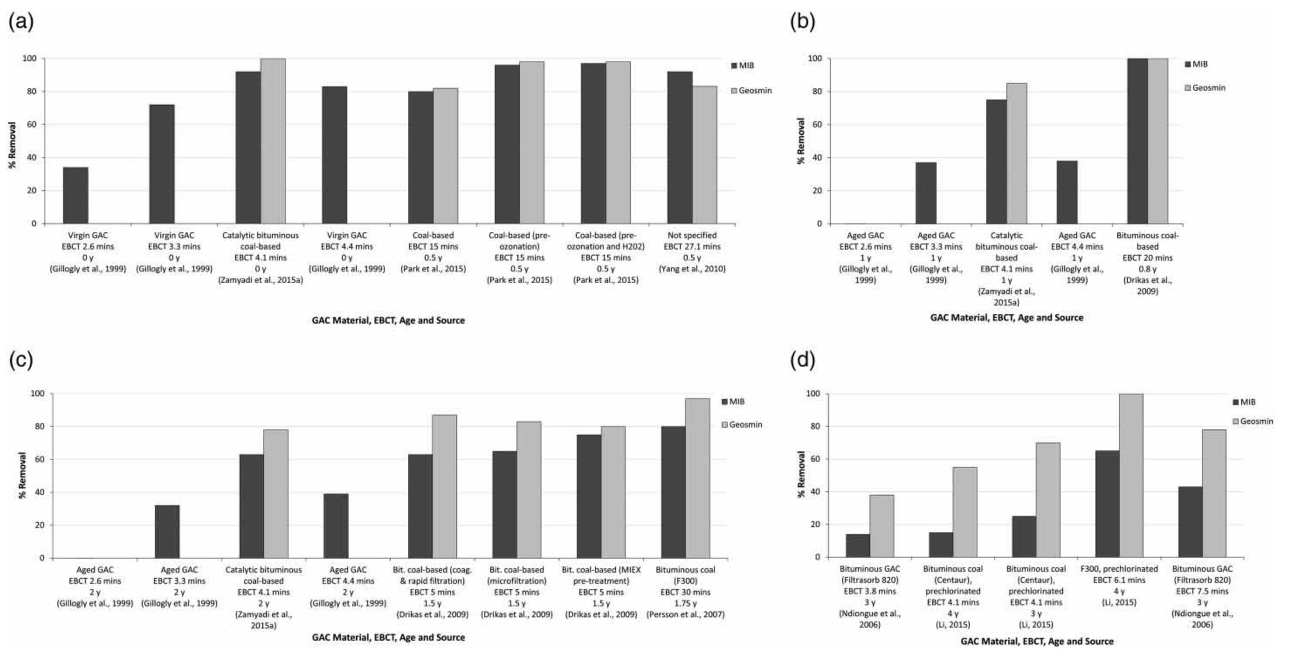

Figure 7 | Impact of EBCT on MIB and geosmin removal by GAC aged (a) less than 6-months-old, (b) 6 months to 1 year, (c) 1 year to 2 years and (d) 2 years to 4 years.

However, the difference between Yang *et al.* (2010) and Park *et al.* (2015) data is attributed to variation in NOM loading due to pre-treatment methods since the water used by Yang *et al.* (2010) only received conventional treatment before filtration through the GAC column. These findings are in accordance with a Zhu *et al.* (2010) publication emphasising the impact of operational factors on microbial community shift and biofilm capacity to remove target compounds.

Figure 7(c) also shows the impact of pre-treatment, with three GACs tested by Drikas *et al.* (2009) varying in removal efficiency. A 1.5-year-old bituminous coal-based GAC pre-treated with MIEX resin achieved a higher MIB removal efficiency than GACs of the same age and EBCT which were pre-treated with microfiltration and conventional treatment. This was due to the higher removal of DOC attained by MIEX treatment in comparison to microfiltration and conventional treatment (Drikas *et al.* 2009).

Figure 7(b) shows an increase in removal efficiency with longer EBCTs among GACs with EBCTs of 3.3 min, 4.1 min, and 20 min. Interestingly, 1- and 2-year-old GACs, each with an EBCT of 2.6 min, achieved negative removal efficiencies of −24% and −45%, respectively, as seen in Figure 7(b) and 7(c). The author attributed this to desorption of MIB taking place after a period of high MIB concentration in the influent water (Gillogly *et al.* 1999). Figure 7(d) also shows the impact of age on GAC removal efficiency with a 3-year-old bituminous coal with an EBCT of 4.1 min achieving a higher MIB and geosmin removal than a 4-year-old bituminous coal with the same EBCT.

Despite being younger and having a longer EBCT, a 3-year-old GAC with an EBCT of 7.5 min achieved a lower MIB and geosmin removal than a 4-year-old pre-chlorinated GAC with and EBCT of 6.1 min, both shown in Figure 7(d). This can again be attributed to a difference in NOM loading since pre-treatment for TOC was not undertaken prior to filtration through the 3-year-old GAC while the 4-year-old GAC had been exposed to H_2O_2 which the author noted improved removal capacity of the GAC (Li 2015).

In contrast to the clear impact of EBCT on GAC removal efficiencies, Figure 8 shows that EBCT had little effect on the BACs tested by existing pilot-studies. Drikas *et al.* (2009) examined the effect of pre-treatment on T&O removal, comparing the impact of coagulation and rapid filtration, microfiltration, and MIEX resin pre-treatment. Each BAC which utilised these different pre-treatment methods achieved both geosmin and MIB removals of over 99% and are shown in Figure 8 under the same bituminous coal-based BAC.

Furthermore, while assessing biofilms on sand media, McDowall (2008) determined that in an EBCT range of 10 min to 30 min, an increase in EBCT did not impact removal efficiency of biofilm. Figure 8 does show that above 99% removal efficiency was achieved by most BACs utilising an EBCT between 5 and 15 min, with Scharf *et al.* (2010) being the only exception; biofilm was only assumed to exist on this GAC and not confirmed, with a lower biomass possibly resulting in these observations.

A 6-year-old full-scale BAC with pre-ozonation, examined by Nerenberg *et al.* (2000), was the oldest BAC studied in the searched literature. Due to its age, the authors assumed its adsorption capacity to be exhausted. The overall average MIB removal efficiency of 91.8% was therefore assumed to be due to only ozonation and biofiltration. This study used an EBCT of 17 min and demonstrated that aged BACs with high EBCTs are capable of removing a

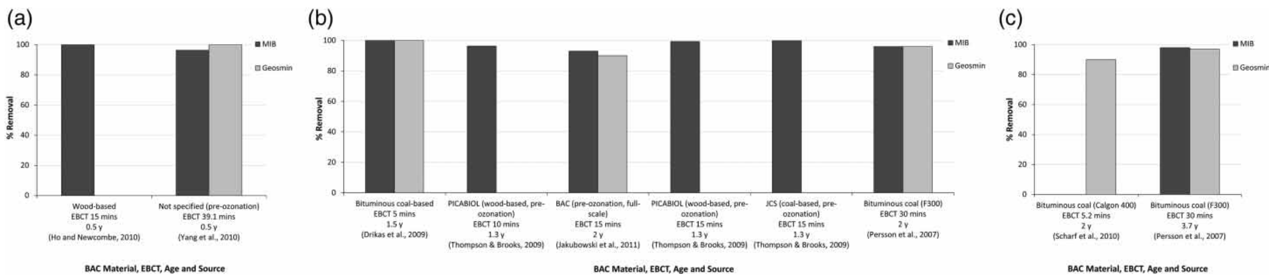

Figure 8 | Impact of EBCT on MIB and geosmin removal by BAC aged (a) less than 6-months-old, (b) 1 year to 2 years and (c) 2 years to 4 years.

sufficient amount of T&O compounds. However, modelling biological activity would be necessary to design and evaluate performance of BAC contactors in full-scale plants in different climate zones. While models have been developed to predict the performance of GAC contactors, no tools have yet been developed to predict the performance and service life of BAC contactors. Further research is required to better understand the establishment of biofilm on carbon and model its performance.

CONCLUSIONS

- GAC is one of the most efficient substance for removal of geosmin and MIB through adsorption. However, its removal efficiency and capacity are heavily influenced by factors including GAC age, EBCT utilised, and loading of NOM on the GAC surface as well as NOM concentration in the influent water. As GAC ages, its ability to achieve high T&O removal efficiencies decreases resulting in contactors needing to be renewed frequently, though longer EBCTs do achieve higher removal efficiencies. High concentrations of NOM also result in lower adsorption rates of T&O compounds.
- BAC and systems utilising both ozonation and biofiltration have achieved higher T&O removal rates than GAC as they involve biodegradation as well as adsorption. BAC can overcome the limitations surrounding use of GAC such as EBCT and age, as 6-year-old BAC contactors can continue to remove over 90% of T&O compounds while the same removal efficiency can only be attained by virgin GACs.
- Although biological activity tests are effective at determining the presence of biofilm, they do not measure the capacity of the biofilm to degrade MIB and geosmin. Rather, as the degradation of MIB and geosmin is determined by specific bacteria and their ability to form a biofilm on the media, targeted biological activity measurement technique are required to accurately determine biofilm MIB and geosmin degradation capacity. Further research is required on modelling the performance of BAC for T&O removal, as well as the full-scale performance of both GAC and BAC as current data is limited. There is also a need for developing an accurate tool to predict the service life of GAC and BAC for T&O removal.

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