

Geosmin and 2-methylisoborneol removal in drinking water treatment

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ABSTRACT

Geosmin (GSM) and 2-methylisoborneol (2-MIB) are metabolites derived from cyanobacteria which produce taste and odor problems in drinking water treatment plants. Conventional treatment processes in water treatment are ineffective for the removal of 2-MIB/GSM. In this study, GSM and 2-MIB doses were applied to the raw water samples. Three powdered activated carbons (PAC), coconut shell, wood, and bituminous, and two oxidants, hydrogen peroxide and potassium permanganate, were evaluated. In addition, all the tests were made with and without flocculation aid (polyacrylamide). All the PACs evaluated showed better GSM and 2-MIB removals when conventional treatment was carried out with the addition of polyacrylamide as a flocculation aid. However, concentrations below the odor threshold were not reached. Regarding the oxidants, hydrogen peroxide presented the highest efficiency in eliminating GSM and 2-MIB when there is no presence of polyacrylamide.

Key words: hydrogen peroxide, polyacrylamide, powdered activated carbon, taste and odor compounds

HIGHLIGHTS

- The removal of odor compounds in drinking water with powdered activated carbon and two oxidants was studied.
- The addition of polyacrylamide as a flocculation aid improved the GSM/MIB removal with the different types of PAC evaluated and the potassium permanganate.
- GSM/MIB concentrations below the odor threshold were not reached in the tests.

INTRODUCTION

Taste and odor problems in water have increased over time and impact the supply of drinking water to communities. The presence of precursor compounds of taste and odor in the water makes users question the safety of drinking water and cause discomfort causing an increase in complaints received by the company providing the service. Suffet *et al.* (1996) reported that 377 utilities in the United States and Canada, equivalent to 16% of the companies surveyed, had serious taste and odor problems, and 43% of these experienced a serious event that lasted more than 1 week. In China, it was reported that 80% of 111 drinking water treatment plants evaluated presented odor problems in raw water, however, the occurrence rate dropped to 45% in treated water (Sun *et al.* 2014).

The treatment and control of taste and odor compounds have become a difficult task, since their odor threshold is very low, generally in the ng/l range. Among the most common odor and taste generating compounds in water are 2-methylisoborneol (2-MIB) and geosmin (GSM) (Antonopoulou *et al.* 2014; Li *et al.* 2019), which are metabolites generated by a wide variety of cyanobacteria in aquatic environments (Watson 2010). These are detectable in low concentrations due to their low odor threshold by humans and are difficult to remove by conventional purification systems (Srinivasan & Sorial 2011). When algae and bacteria blooms occur in water sources, the concentrations of GSM and 2-MIB can be above the odor threshold, which can range from 4 to 10 ng/l for GSM and 15 to 29 ng/l for 2-MIB (Watson *et al.* 2008; Antonopoulou *et al.* 2014).

GSM and 2-MIB are compounds that are difficult to remove with conventional water treatment processes, such as coagulation, sedimentation, and filtration; only advanced water treatment processes can be considered as alternative methods to remove GSM and 2-MIB. The processes reported by different authors include oxidation (Park *et al.* 2006), adsorption of activated carbon (Cook *et al.* 2001; Zamyadi *et al.* 2015a; Bong *et al.* 2021),

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nanofiltration (Dixon *et al.* 2011), and ultraviolet rays with H₂O₂ (or TiO₂) (Rosenfeldt *et al.* 2005; Zamyadi *et al.* 2015b).

The drinking water treatment company in Bucaramanga, Colombia, uses conventional treatment, in which coagulation is the beginning of this process. However, when raw water conditions require it, it is common to find pretreatment units for the solids removal or other unusual contaminants. In addition, conventional coagulants, including aluminum and iron salts, and flocculation aids are widely used in water treatment facilities.

The Bosconia Drinking Water Treatment Plant (DWTP-B) of Bucaramanga presents flavor events in the drinking and raw water caused by the growth of periphytic algae, in dry seasons, when the levels in the Surata River drop. The perception of smell and taste in water is attributed to different factors, such as the presence of inorganic compounds like metallic ions contributed by anthropogenic activities (mining, industrial discharges) and organic compounds of natural or synthetic origin, contributed by domestic wastewater discharges or wastewater from agricultural and livestock activities. However, in previous studies carried out by the aqueduct company, the presence of cyanobacteria has been identified in times of low flows in the Surata River, which are associated with flavor water events (Ortenberg & Telsch 2003).

When there are taste problems in the DWTP-B, coconut shell activated carbon at the flocculation stage and polyacrylamide in the pretreatment are added, obtaining good results on some occasions, being necessary to evaluate different alternatives to carry out in new events of bad taste. Among the alternatives for the control of taste precursor substances in water is the use of oxidants. Hydrogen peroxide has been used as an environmentally friendly algacide, due to it does not produce chemical residues in the water (Yang *et al.* 2018). On the other hand, potassium permanganate has been used to control algae and the associated metabolites (Ho *et al.* 2009), relating it to better functioning of the coagulation–flocculation process (Chen *et al.* 2009).

The goal of this study was to assess the powdered activated carbon and the oxidation with hydrogen peroxide and potassium permanganate as alternatives for removing the taste of water, caused by the presence of GSM and 2-MIB in the DWTP-B.

METHODS

Water source and sampling points

This study was performed in the DWTP-B located on the Matanza-Bucaramanga road (Colombia). This DWTP is supplied by the Surata River and is divided into the following stages: pretreatment (Parshall flume, grit removal, and pre-settler), rapid mixing, flocculation, clarification, rapid filtration, and disinfection with chlorine. The treatment capacity is 2,000 l/s. Aluminum sulfate is used as coagulant and, in some cases, polyacrylamide is used as flocculation aid. Raw surface water used in this study was collected from the water intake on the Surata River. Later, the water samples were taken to the drinking water laboratory at the Pontificia Bolivariana University, where the experimental tests were carried out. The characteristics of the raw water used in the tests were: turbidity 33.5 ± 5.5 NTU; pH 8.0 ± 0.1 units; conductivity 177.4 ± 28.8 μ S/cm and color 60.0 ± 10.1 PCU, dissolved organic carbon (DOC) 3.0 and 2.9 mg/l for the conventional treatment without and with flocculation aid, respectively, indicating little presence of dissolved organic matter.

Bench-scale experiments

The dates on which the water samples from the DWTP-B were collected were characterized by no algal blooms in the river. Therefore, it was necessary to add concentrations of GSM and 2-MIB with patterns to simulate the presence of odorous compounds. Two combinations of GSM and 2-MIB were used by adding the stock solution to raw water: C1: 114 ng/l GSM – 45 ng/l 2-MIB for treatment without flocculation aid and C2: 24 ng/l GSM – 968 ng/l 2-MIB for treatment with flocculation aid. The flocculation aid was added when there was a predominance of 2-MIB in the combination used, that is, in C2.

All the tests were performed simulating the conventional drinking water carried out in the DWTP-B through the jar-test using equipment PHIPPS and BIRD. The following conditions were used to simulate the conventional drinking water treatment: rapid mixing at 300 rpm for 1 min, flocculation at 34, 23, 24, and 14 rpm for 6 min each speed and settling for 34 min. The settled samples were passed through an ascending filter and finally disinfected with a contact time of 30 min. Tests were carried out on 2.0 L water samples and the coagulants were added to the jars immediately after the start of rapid mixing. Disinfected water samples were sent to the laboratory for the measurement of GSM and 2-MIB concentrations.

When there are taste problems in the DWTP-B, coconut shell activated carbon is added at the flocculation stage at doses up to 4 mg/l and the polyacrylamide addition in the pretreatment. For this reason, tests were initially carried out with coconut shell powdered activated carbon (CS-PAC), changing the doses (2, 5, and 8 mg/l), the application point (pretreatment and flocculation), and the flocculation aid addition in the pretreatment.

Besides, tests with wood powdered activated carbon (W-PAC) and bituminous powdered activated carbon (B-PAC) were carried out to compare with CS-PAC. These tests were performed with and without polyacrylamide which was applied in the rapid mixing that simulates the pretreatment Parshall flume.

In addition, two oxidants were assessed: hydrogen peroxide (HP) and potassium permanganate (PP) with two doses each (0.5 and 1.5 mg/l). The tests were carried out simulating the point of application of the oxidants in the pretreatment Parshall flume.

Chemical products

The 2-MIB (98%) and GSM (97%) were obtained as solids from Sigma–Aldrich Co. A stock solution of 500 mg of GSM per mL of pure water was prepared and stored at 4 °C. The coagulant used was aluminum sulfate with 7% of alumina and the flocculation aid was a non-ionic ultra-molecular weight polyacrylamide. The PACs and the oxidants were supplied by the Bucaramanga water supply company.

Analytical methods

The turbidity was measured using a Turbidimeter (Marca HACH 2100N, USA), and the pH and conductivity were measured using a multiparameter (Marca Fisher Scientific, Ottawa, ON, Canada). The color and the DOC were measured according to the standard methods (APHA *et al.* 2017).

Samples for 2-MIB and GSM analysis were collected headspace-free in 45 mL acid-washed bottles with caps lined with PTFE septa for subsequent analysis. The simultaneous extraction–concentration of 2-MIB and GSM from aqueous samples was carried out using a fused silica fiber coated with PDMS/DVB 65 µm thick (PDMS/DVB-65 µm *Stableflex*). The separation, detection, and quantification of GSM and 2-MIB were carried out by using a gas chromatograph (Agilent 7890B) with a mass spectrometer detector (Agilent 5977A) coupled to a purge and trap (Tekmar) sample induction system using the selective ion monitoring mode (GC-MS/SIM). 2-MIB and GSM were used as the internal standard. The method detection limit (MDL) was 1 ng/l for GSM and 3 ng/l for MIB, respectively. Before analysis, samples were filtered through a 0.45 mm PTFE filter.

RESULTS AND DISCUSSION

The results of the physicochemical parameters obtained in the tests show good removal of turbidity and color, achieving final values that comply with the Colombian regulations for drinking water (turbidity < 2 NTU, color < 15 PCU, pH between 6.5 and 9.0 units, and conductivity < 1,000 µs/cm).

Powdered activated carbon

The results of GSM removal when CS-PAC was added in the pretreatment and flocculation stage are between 15 and 58% (Figures 1 and 2). Figures 1 and 2 show the GSM removal reached with the treatment without and with

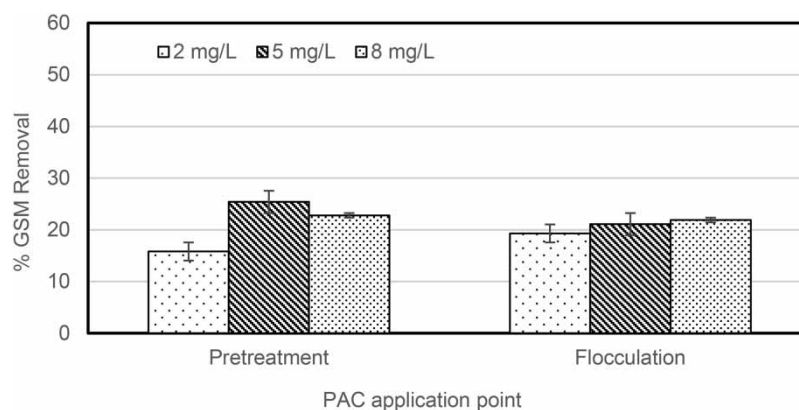


Figure 1 | GSM removal with CS-PAC and conventional treatment without polyacrylamide.

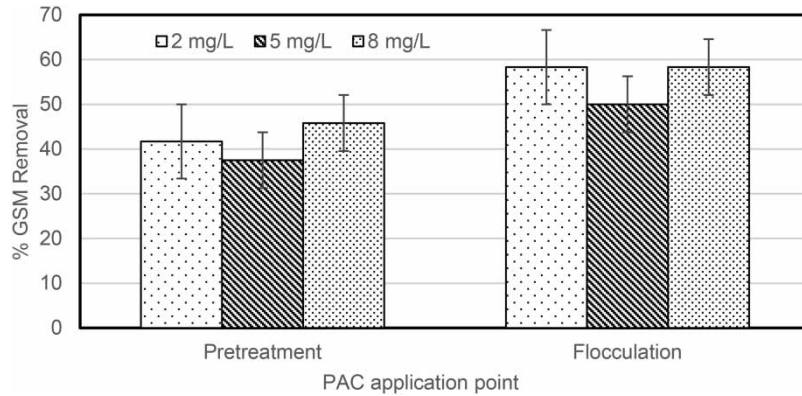


Figure 2 | GSM removal with CS-PAC and conventional treatment with polyacrylamide.

the addition of polyacrylamide, respectively. According to the results, the removal reached with the treatment without the addition of polyacrylamide was between 15 and 25% (Figure 1). In addition, the values obtained for the remaining GSM, regardless of the application point or the CS-PAC dose applied are high (90 ± 3.7 ng/l) and exceed the reported values of the odor threshold. The highest removals were obtained when CS-PAC was applied in the treatment with the addition of polyacrylamide in the pretreatment, varying between 38 and 58% (Figure 2). The GSM concentration remaining after treatment was lower (12.3 ± 2.1 ng/l), however, it was higher than the odor threshold.

The 2-MIB removal results obtained by dosing CS-PAC in the pretreatment and the flocculation stage are shown in Figures 3 and 4. Initially, the removals achieved with the treatment without the addition of polyacrylamide are shown in Figure 3 and then the values for polyacrylamide treatment are shown in Figure 4. According to the results, the removals obtained when the treatment without the addition of polyacrylamide was used were between 11 and 22% and there were no significant differences between the CS-PAC doses or between the application

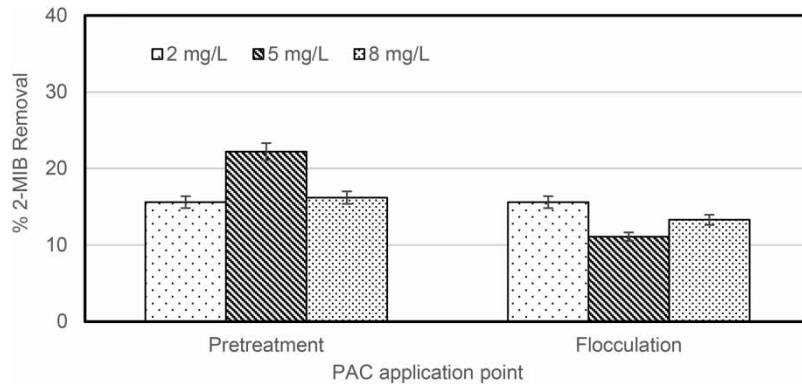


Figure 3 | 2-MIB removal with CS-PAC and conventional treatment without polyacrylamide.

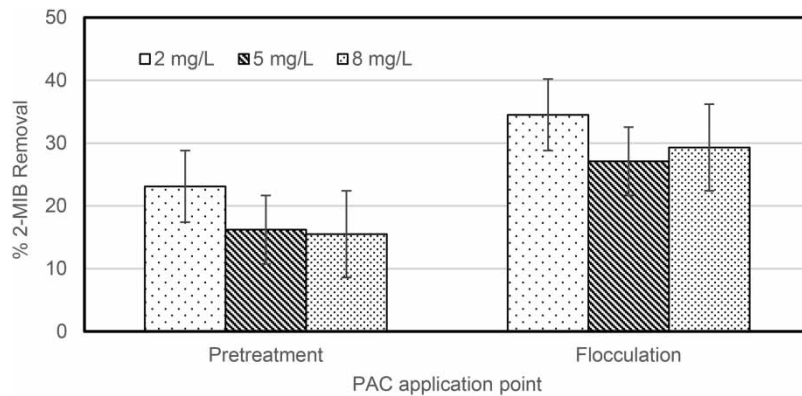


Figure 4 | 2-MIB removal with CS-PAC and conventional treatment with polyacrylamide.

points. The values obtained for the remaining 2-MIB, regardless of the point of application or the CS-PAC dose applied, are high and exceed the reported values of the odor threshold (2-MIB remaining: 38 ± 1.7 ng/l).

In the conventional treatment with the addition of polyacrylamide, the 2-MIB removals varied between 15 and 35%. In this case, significant differences were observed between the applied CS-PAC doses and between the application points, achieving higher removals when the CS-PAC was applied in the flocculation stage. Although the 2-MIB removal in these tests was higher, the remaining 2-MIB concentration was very high (732.8 ± 72.6 ng/l), exceeding the odor threshold. This is because the 2-MIB concentration applied to the raw water sample was higher. In all tests, 2-MIB was not removed below the odor threshold with the doses and application points evaluated, confirming that CS-PAC is not the best alternative to use. The quantitative results of GSM and 2-MIB showed that doses of 2, 5, and 8 mg/l are not optimal to reach removals up to the odor threshold. Kim *et al.* (2014) reported that to achieve sufficient removal of 2-MIB (50 ng/l) and GSM (200 ng/l), powdered activated carbon should be added at concentrations of 18 and 20 mg/l, respectively. In addition, the reduction of the adsorption capacity of activated carbon, due to the presence of dissolved natural organic matter (NOM) is another problem that arises in the elimination of these metabolites (Zoschke *et al.* 2011). Typical concentrations of NOM in drinking water sources are in the range of 2–10 mg/l DOC, while concentrations of micro-contaminants are in the range of 10–1,000 ng/l. In this case, the DOC presented values of 3.0 mg/l. In general, two mechanisms are described to explain the reduction in adsorption capacity by NOM: (a) larger NOM molecules can block activated carbon pores and restrict the access of smaller molecules and (b) NOM molecules can compete directly with micro-contamination for adsorption sites. However, GSM is much more absorbable than the 2-MIB in the NOM-water containing, as it has a lower molecular weight and flatter structure than the 2-MIB, allowing entry into the smallest micropores and, therefore, it has less direct competition with NOM molecules. Though some NOM molecules are small enough to enter the micropores and thus directly compete with GSM and 2-MIB for adsorption sites (Newcombe 2006).

At higher initial concentrations of GSM and 2-MIB, better removals were reached because there is a greater probability of collision between the activated carbon particles and the metabolites. Additionally, it is observed that the use of polymer reports higher removals since the large flocs formed also remove these odoriferous substances from the water by adsorption (Aguilar *et al.* 2005).

The GSM and 2-MIB removals obtained with the different types of PAC are in the range of 8.8–62% (Figures 5–8).

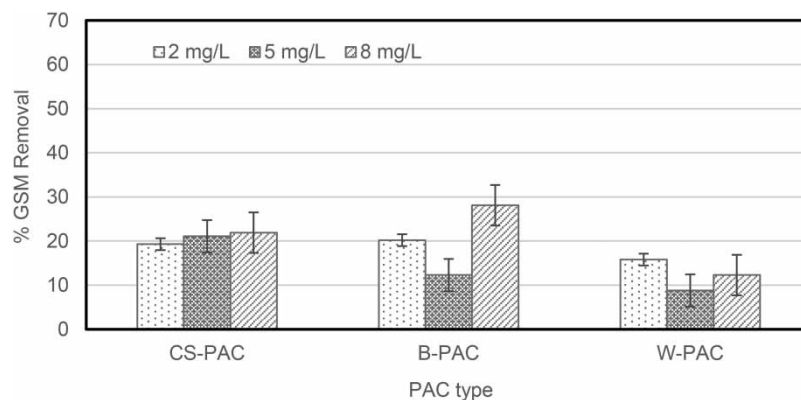


Figure 5 | GSM removal with different PACs and treatment without polyacrylamide.

According to the results, GSM removals achieved with the treatment without the addition of polyacrylamide were between 8.8 and 28.1% and there were no significant differences between the PAC types or between the doses applied. While in the treatment with the addition of polyacrylamide, the removals varied between 33.3 and 62.5% and there were no significant differences between the doses of PAC applied, but there were between the PAC types, reaching higher removals with CS-PAC and W-PAC. The addition of polyacrylamide could increase the sedimentation rate of the formed flocs, as has been reported by some authors (Aguilar *et al.* 2005). However, the values obtained for the remaining GSM, regardless of the type of PAC applied, are high (93.8 ± 6.8 ng/l and 12.1 ± 2.3 ng/l, without and with flocculation aid, respectively) and exceed the reported threshold values of taste. As for the 2-MIB, in the conventional treatment without the addition of polyacrylamide,

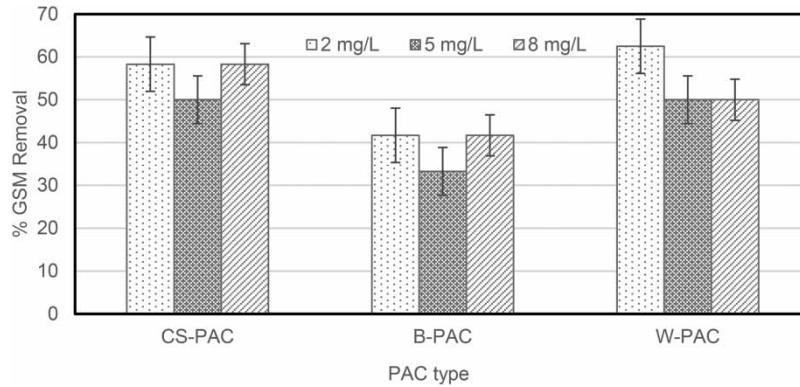


Figure 6 | GSM removal with different PACs and treatment with polyacrylamide.

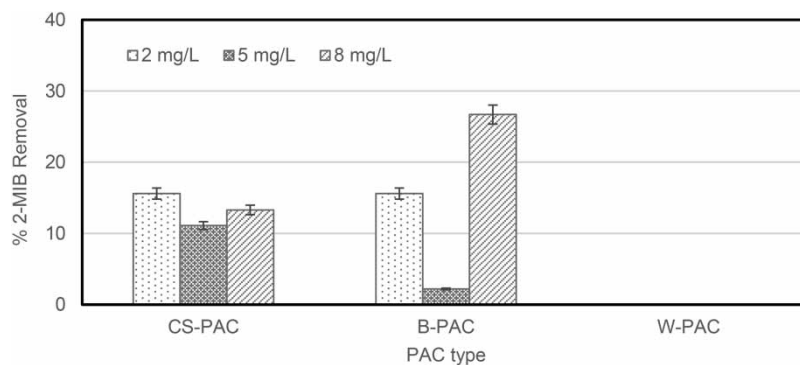


Figure 7 | 2-MIB removal with different PACs and treatment without polyacrylamide.

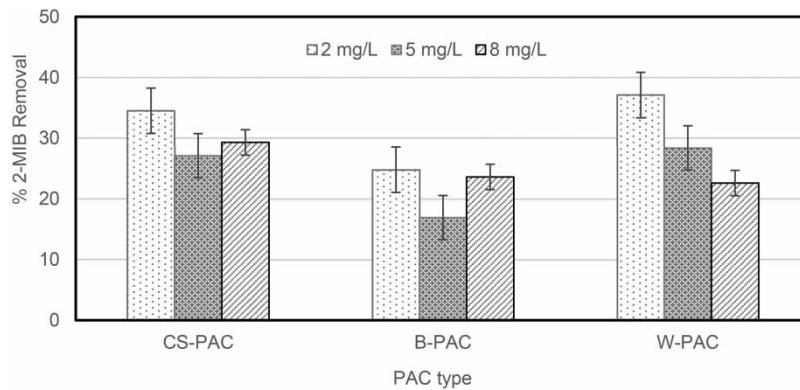


Figure 8 | 2-MIB removal with different PACs and treatment with polyacrylamide.

very low removals were achieved with a maximum value of 26.7%; furthermore, W-PAC did not show any removal. In the treatment with the addition of polyacrylamide, greater removals were achieved, showing significant differences between the PAC types; in this case, B-PAC was the one with the lowest 2-MIB removals. The results reflect that the origins of the raw materials of the activated carbons evaluated (coconut shell, wood, and bituminous) are not a decisive factor in the adsorption of GSM and 2-MIB when the conventional treatment is carried out without the addition of polyacrylamide, however, when it is added as a flocculation aid, the type of PAC influences the removal. *Yu et al. (2007)* evaluated three PAC types, bituminous (B1–B2), fruit peel (F), and wood (W), with fruit peel-based carbon having the highest adsorption capacity than wood-based and bituminous carbons, mainly attributing this to their larger micropore volumes. *Li et al. (2019)* found that 2-MIB can be found

dissolved or bound and the removal rate of this compound increased with the concentration in the raw water. A high concentration of 2-MIB in raw water corresponds to a high proportion of bound 2-MIB, which can be removed by sedimentation or filtration. Therefore, conventional treatment can reasonably effectively remove 2-MIB at higher concentrations.

Oxidants

The removals achieved with the oxidants used are presented in Figures 9 and 10. In the conventional treatment without the addition of polyacrylamide, removals of GSM and 2-MIB greater than 80% were achieved using hydrogen peroxide for their elimination. However, by adding polyacrylamide as a flocculation aid, removals with this oxidant drop to less than 10%.

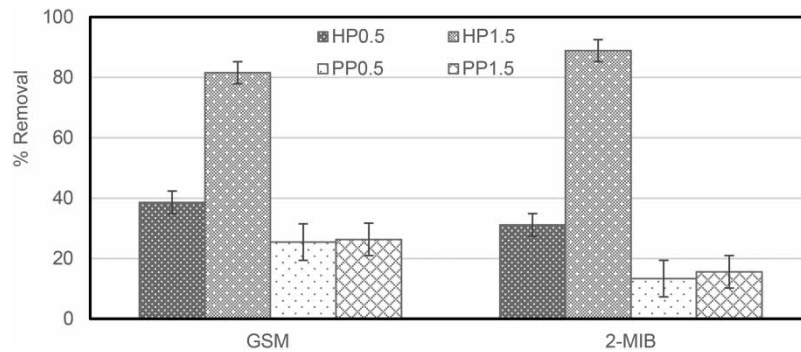


Figure 9 | GSM and 2-MIB removal with oxidants and treatment without polyacrylamide.

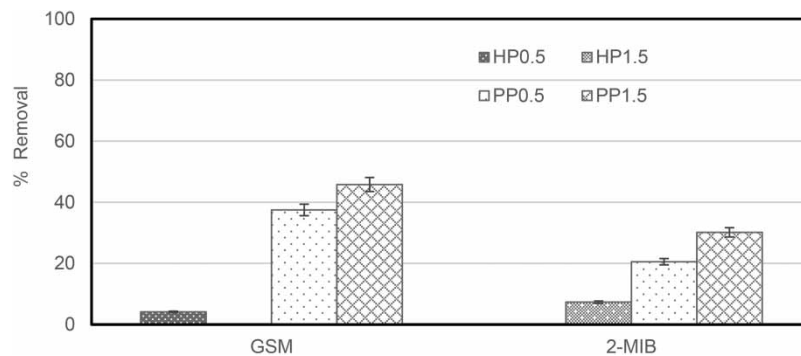


Figure 10 | GSM and 2-MIB removal with oxidants and treatment with polyacrylamide.

The results of the evaluation of the oxidative capacity of potassium permanganate and hydrogen peroxide are promising concerning the evaluation of activated carbon. Hydrogen peroxide was able to oxidize up to the odor threshold with the maximum dose (1.5 mg/l), when using only the conventional treatment without the application of polymer in the treatment. However, any residual oxidant must be fully removed before the disinfection process, as it exerts a strong chlorine demand and could compromise final disinfection, leading to insufficient chlorine residual within the distribution system (Zamyadi *et al.* 2015b). Xu *et al.* (2019) found that the concentration of 2-MIB decreased with the application of doses between 3.0 and 20.0 mg/l H_2O_2 after 8 h of reaction, showing that the capacity of H_2O_2 to eliminate 2-MIB was greater than that of $CuSO_4$ and $KMnO_4$. This can be attributed to the higher oxidation capacity of H_2O_2 that can produce highly reactive hydroxyl radicals and to the higher dose of H_2O_2 in this study.

Potassium permanganate showed better results in the treatment with the addition of polyacrylamide. According to Chen & Yeh (2005), potassium permanganate promotes the aggregation of algal cells and is related to better functioning of the coagulation, flocculation, and sedimentation processes (Chen *et al.* 2009). Furthermore, when $KMnO_4$ is introduced, organic matter is oxidized to lower molecular weight fractions, producing inorganic carbon and a higher concentration of oxygenated functional groups, which contribute to organic matter binding

to coagulants, leading to a reduction in the coagulant dose and an increase in the elimination of organic matter by coagulation (Naceradska *et al.* 2017). The addition of polyacrylamide allows for better contact between the oxidized organic matter and the coagulant, forming flocs that can be removed during sedimentation. One of the drawbacks of the application of oxidants is the increase in the total extracellular concentration of odorous and/or toxic metabolites if cell damage and metabolite release exceed the rate of oxidation during drinking water treatment (Wert *et al.* 2014).

CONCLUSION

The CS-PAC presented better removals of GSM and 2-MIB when conventional treatment was carried out with the addition of polyacrylamide as a flocculation aid. However, concentrations below the odor threshold were not reached.

The application of different types of PAC confirmed the positive effect of the addition of polyacrylamide as a flocculation assistant since better removals were achieved with all the PACs evaluated, both for GSM and 2-MIB.

Hydrogen peroxide presented the highest efficiency in removing flavor precursors when there is no presence of the polymer (polyacrylamide), however, when applying this polymer, potassium permanganate increases its effectiveness in removing GSM and 2-MIB of water.

Although good removal of odorous substances was obtained in the experiments, it is necessary to investigate other removal methods such as nanofiltration membranes or advanced oxidation processes, as well as the combination of these with adsorption on activated carbon.

ACKNOWLEDGEMENTS

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DATA AVAILABILITY STATEMENT

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

CONFLICT OF INTEREST

The authors declare there is no conflict.

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