Application of double filtration with activated carbon for the removal of phenols in drinking water treatment processes

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ABSTRACT

The primary source of the discharge of phenols into the environment is industrial activity, such as the production of pharmaceuticals, plastics, and pesticides, being the majority discharged into surface water sources, reaching concentrations around 0.001 to 0.400 mg/L. These compounds are considered a priority contaminant due to their toxicity to aquatic life and effects on human health. The presence of phenols even at low concentrations generates flavour and odour in drinking water. Due to the molecular stability and solubility of phenols in water, their removal by conventional water treatment methods is inefficient. However, adsorption with granular activated carbon (GAC), after conventional filtration with sand and anthracite, is an efficient technique for the reduction of organic compounds such as phenols. This paper studied the effect of applying double filtration to the reduction of phenols present in the filtered water of a conventional drinking water treatment plant, using two types of GAC (vegetable and mineral) and three GAC:Sand configurations (100:00; 00:100; 50:50). The configurations with GAC showed an efficient reduction of turbidity, organic matter indicator variables (UV_{254} absorbance and total organic carbon) and phenols, the mineral GAC being the most efficient GAC.

Key words | adsorption, double filtration, drinking water, filter media, granular activated carbon (GAC), phenols

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INTRODUCTION

Water is often the main route of human exposure to pathogens, toxins, and organic and inorganic contaminants. In Colombia, around 60% of drinking water treatment plants (DWTPs) are conventional and supplied by surface water source, that generally presents high turbidity levels, reaching levels in the order to 3,000 UNT (Pérez-Vidal *et al.* 2016); in addition, the sources are exposed also to contaminants such as phenols.

The presence of phenols in the environment is mainly a consequence of human activities (industrialization and agricultural activities). Wastewater from industrial processes (for the production of pharmaceuticals, perfumes, explosives, phenolic resins, plastics, textiles, petroleum, dyes, leather, paper and pesticides and in cokers and tar distilleries) contains high concentrations of phenols, of which approximately 73.3% reach water, approximately 26.3% reach the air and approximately 0.4% reach soil and aquatic sediments (Mohamed *et al.* 2005).

The average concentration of phenols in natural waters varies from one type of water to another; in contaminated natural water, levels can range from 0.001 to 0.400 mg/L and even greater concentrations (Breton *et al.* 2003; Pérez-Vidal *et al.* 2016). Phenols and their compounds have been classified as priority toxic pollutants by the Environmental Protection Agency, because their presence in drinking water bodies represents a high risk to consumers (EPA 2014).

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In drinking water, phenols generate odour and taste and may have negative effects on the environment and human health (Mohamed et al. 2005). The World Health Organization (WHO 1963) strictly regulates phenols and has established a limit of 0.001 mg/L in drinking water; in Colombia, levels cannot exceed 0.002 mg/L in surface water (Ministry of Agriculture 1984) and 0.001 mg/L in drinking water (NTC 813 2004). However, conventional or full-cycle DWTPs, which are widely used in water purification, achieve a limited removal of phenols, less than 10%, due to the stability and solubility of these compounds in water (Kim & Kang 2008).

For the removal of phenols and other forms of organic matter, in addition to activated carbon powder, filters with adsorbent media like granular activated carbon (GAC) are used to replace the granular media commonly used in rapid filters (sand or sand and anthracite) or as a step after conventional filtration because the properties of activated carbon, such as surface area, porosity and surface chemistry, provide a higher capacity for the adsorption of organic molecules such as phenols (Aksu & Kabasakal 2004). Authors including Kim & Kang (2008) and Tan et al. (2013) have shown the efficiency of dual sand and GAC media in removing organic compounds, in which the sand located at the bottom of the filter is responsible for the polishing step (Perea et al. 2013).

This work evaluated the influence of two types of GAC (vegetable and mineral) as a second filter medium in the removal of phenols present in filtered water (filtered with sand and anthracite) from a DWTP supplied by the Cauca River (Cali, Colombia), which was doped with two concentrations of phenols, close to 0.30 and 1.0 mg/L, before the double filtration with GAC. Taking into account Colombia's drinking water standards, one of the most important quality parameters is turbidity, so in this study, in addition to measuring the absorbance of UV_{254}, total organic carbon (TOC) and phenols, this variable was also measured.

METHODS

Preparation of doped water with phenols

Water filtered through sand and anthracite filters from a conventional DWTP supplied by the Cauca River, which treats water at a flow rate of 6.6 m^3/s, was used. The filtered water was doped with a phenol concentration close to the maximum value found in the Cauca River (0.3 mg/L, Pérez-Vidal et al. 2016) and a higher dose of approximately 1 mg/L and subjected to a second filtration with GAC. The phenol solutions used in the tests were prepared from a standard with a concentration of 1,000 mg/L; 36 and 120 mL of this phenolic solution were diluted in 120 L of filtered water to obtain concentrations close to 0.30 and 1.0 mg/L.

Turbidity (NTU) (2130B) (Turbidimeter 2100N, HACH, Colorado, USA), UV_{254} absorbance (cm^{-1}) (2510B) (Spectrophotometer DR5000, HACH), TOC (mg/L) (5310B) (TOC-VCPH, Shimadzu, Kyoto, Japan) and phenols (mg/L) (5530B) (UV-VIS Spectrophotometer UV-1800, Shimadzu) were measured in the phenol-doped filtered water from the DWTP and the second filtration effluent according to protocols of the American Public Health Association (APHA 2012).

Experimental unit

Transparent glass columns 40 cm long, with a nominal diameter of 25 mm, an internal diameter of 19 mm and a 15 cm filter bed height were used; ten experimental units were used for each doped phenol concentration (Figure 1). The assay was carried out for 7 hours; a flow distribution system was used to allow the transfer of the filtered phenol-doped water to each experimental unit during the assay (Figure 1). The flow rate was 12 mL/min for a constant filtration rate of 2.54 m/h. The empty bed contact time for configurations of 100% GAC was 3.9 min and for 50% GAC was 2 min, acceptable values for laboratory filters (Di Bernardo et al. 2011).

Evaluation of filtration

Three media were used: silica sand, GAC of vegetal origin (VAC) made from coconut husk and GAC of mineral origin (MAC) made from grains of bituminous coal. All of it was of commercial origin. Table 1 shows the characteristics of the adsorbent filter media used.

The five configurations evaluated in duplicate (R) are shown in Table 2, for a total of ten experimental units.

For the turbidity (NTU) and UV_{254} (cm^{-1}) measurements, filtered water samples were taken every 15 minutes.

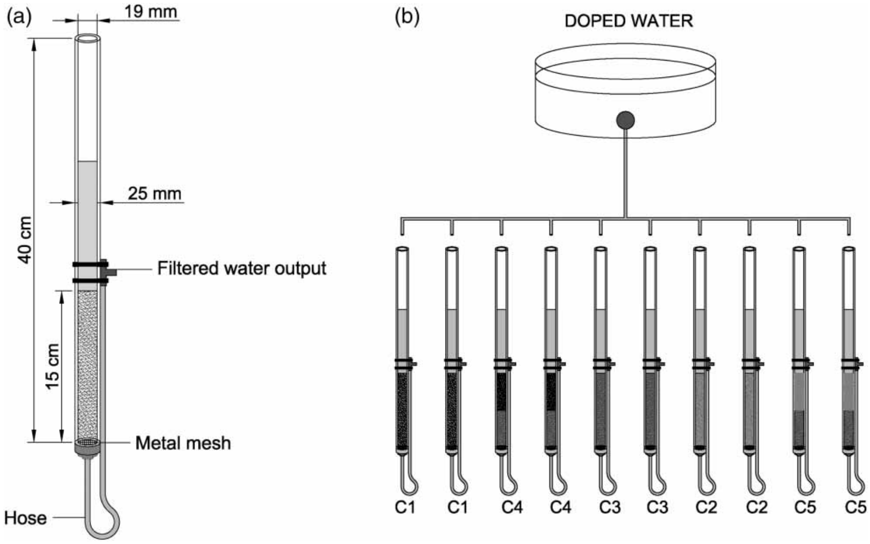

Figure 1 | Experimental unit and filter media configurations.

Table 1 | Characteristics of filter media

	VAC		MAC		
Characteristics	**Min**	**Max**	**Min**	**Max**	**Sand**
Abrasion number	86	–	75	–	–
Iodine number (mg/g)	900	–	850	–	–
Bulk density (g/cm^3)	0.48	0.58	0.48	–	–
Effective size (mm)	0.8	1.2	0.55	0.75	0.61
Coefficient of uniformity	–	2.1	–	1.9	<1.7

Source: Provider information.

Table 2 | Configurations used for the evaluation of filtration

Configuration	VAC (%)	MAC (%)	Sand (%)	Reference
C1	100	–	–	Huang *et al.* (2007), Kim & Kang (2008) and Gibert *et al.* (2013)
C2	–	100	–	
C3	–	–	100	
C4	50	–	50	Kim & Kang (2008) and Tan *et al.* (2013)
C5	–	50	50	

For the analysis of TOC (mg/L) and phenols (mg/L), the required sample volumes (1 L per sample) were collected at 135, 240, 330 and 420 minutes.

Data analysis

To determine statistical significant differences between the configurations of filter media evaluated, an analysis of variance (ANOVA) was performed (a value of P less than 0.05 indicated significant differences between the evaluated criteria). In addition, Tukey's multiple comparison test was performed using the Minitab 17 statistical program.

RESULTS AND DISCUSSION

Characteristics of the adsorbent media

In relation to the adsorbent media, this had similar characteristics, except for the sources and the effective sizes. The VAC made from coconut shell tends to have small pores, whereas in the manufacture of MAC, a wide range of pores tends to form. Therefore, MAC is usually used for applications in which the compounds to be retained are of different molecular sizes, as in the case of the substances present in water (Gupta & Ali 2013). According to these characteristics, the MAC material could be expected to present a greater reduction of organic compounds. Sand features smaller effective size and coefficient of uniformity than GAC, which is expected for this type of medium and favours the entrapment of particles that lead to turbidity (Friedler & Alfiya 2010; Christopher 2012).

Characteristics of DWTP-filtered water and phenol-doped water

Table 3 shows the characteristics of the filtered water and the water doped with phenols.

All the parameters, except for the turbidity, varied as a function of the addition of phenol at concentrations 1 and 2; this result is because turbidity is associated with the presence of particulate matter, whereas colour is related to the presence of natural metal ions such as iron and manganese, humus, dissolved organic matter, plankton and industrial waste, and the remaining parameters are associated with the presence of organic compounds (Crittenden *et al.* 2012). The results of the parameters that indicate organic matter (UV_{254} values and TOC) increased with increasing concentration of phenols. These parameters also show that the water already contained organic matter such as natural organic matter (NOM) or other organic compounds.

Evaluation of filtration

Turbidity

Figure 2 shows the results for turbidity over time for all the configurations evaluated using concentrations 1 and 2.

In general, it was observed that in all the filter configurations, turbidity values lower than the initial level were reached. The filters with 100% sand showed the best performance, given that between 80% and 100% of the data were lower than the initial level for both concentrations. Regarding the filters that used GAC, it was evident that after 310 minutes, the removal of turbidity decreased for both concentrations. This pattern occurs because the media with GAC reaches capacity more quickly due to the particle retention capacity of GAC, which is related to the effective size of the media, whereas the sand has a smaller size and a greater specific area, which facilitates the removal of turbidity. In this sense, filters in which only GAC is used are more susceptible to the penetration of turbidity (Christopher 2012).

However, the ANOVA and Tukey tests indicated that when comparing the GAC configurations with sand, there were no significant differences, except for with the C4 configuration (50% VAC). Feng *et al.* (2012) state that there is little difference between dual-media filters composed of GAC and sand and sand-only composite filters in terms of turbidity removal. Due to the costs and processes involved, it is not recommended to use GAC with the objective of removing turbidity.

Table 3 | Characterization of filtered water and water doped with phenol concentrations 1 and 2 (mg/L)

Parameter	Filtered water	Doped water [1]	Doped water [2]
Turbidity (NTU)	0.1600	0.1900	0.2300
Real colour (UPC)	6	8	10
UV_{254} (cm^{-1})	0.0280	0.0490	0.0630
TOC (mg/L)	2.7610	3.3680	4.1970
Phenols (mg/L)	0.0094	0.2453	1.1960

UV_{254}

Figure 3 shows the results of the UV_{254} measurements over time for all the configurations evaluated using concentrations 1 and 2.

The GAC filters exhibited better performance in terms of reducing the UV_{254} values, with removal efficiencies of up to 93%, which was confirmed by the statistical analysis results, in which for both concentrations, the 100% sand configuration differs from all the other configurations (<41%). Between the two GAC materials, MAC exhibited worse performance with less variability than VAC. The C4 configuration also showed significant differences from the other configurations due to the lower efficiency of this configuration. Thus, these results validate several studies demonstrating that GAC is one of the materials with the greatest capacity for the adsorption of organic compounds (Crittenden *et al.* 2012; Gupta & Ali 2013).

According to Serrano *et al.* (2015), who used a conventional treatment with sand filters, the UV_{254} values of the filtered water decrease by a low percentage (<18%). Other studies comparing filtration with 100% sand, 100% GAC configurations and dual media consisting of sand and GAC or other materials (Babi *et al.* 2007; Lu *et al.* 2010; Feng *et al.* 2012) found that GAC filters and dual filters

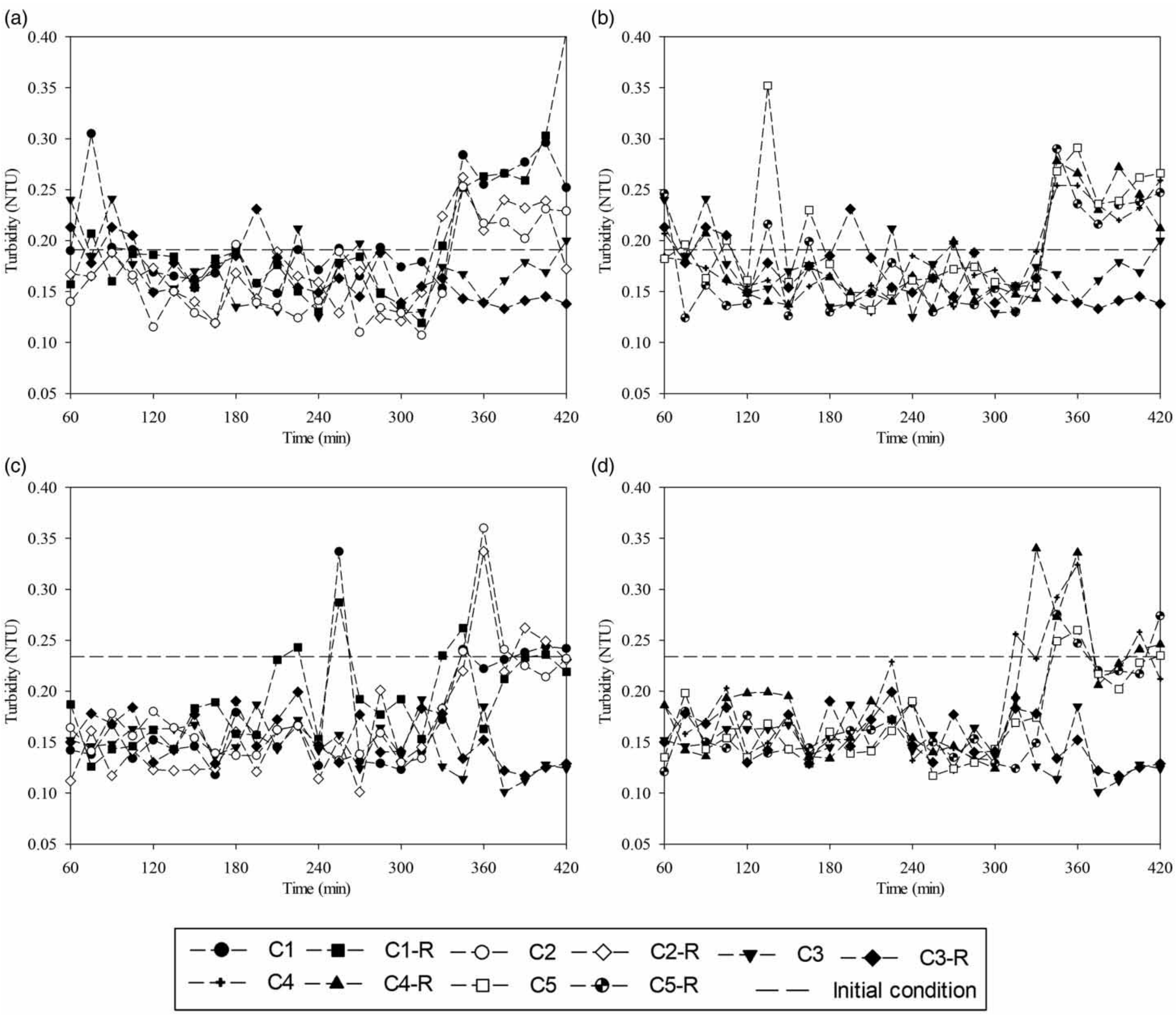

Figure 2 | Turbidity over time for all configurations using phenol concentrations 1 and 2. 1. Phenol at 0.25 mg/L. 2. Phenol at 1.20 mg/L.

have higher efficiency than sand filters in the removal of organic matter, represented by the decrease in UV_{254} absorbance, consistent with this study.

TOC and phenols

Table 4 shows TOC results with the replica (R) over time for phenol concentration 1.

Table 5 shows TOC results with the replica (R) over time for phenol concentration 2.

Table 6 shows phenol results with the replica (R) over time for phenol concentration 1.

Table 7 shows phenol results with the replica (R) over time for phenol concentration 2.

Figure 4 shows the reduction of TOC and phenols for all the configurations used.

GAC has been used with favourable results for the removal of phenols from water (Aksu & Kabasakal 2004) because GAC has an excellent adsorption capacity for relatively low molecular weight organic compounds such as phenols (Mohamed et al. 2005). Figure 4 shows that the 100% GAC configurations presented better results compared to the 50% GAC configurations, and a correlation to the amount of GAC available for the adsorption of organic compounds could be shown.

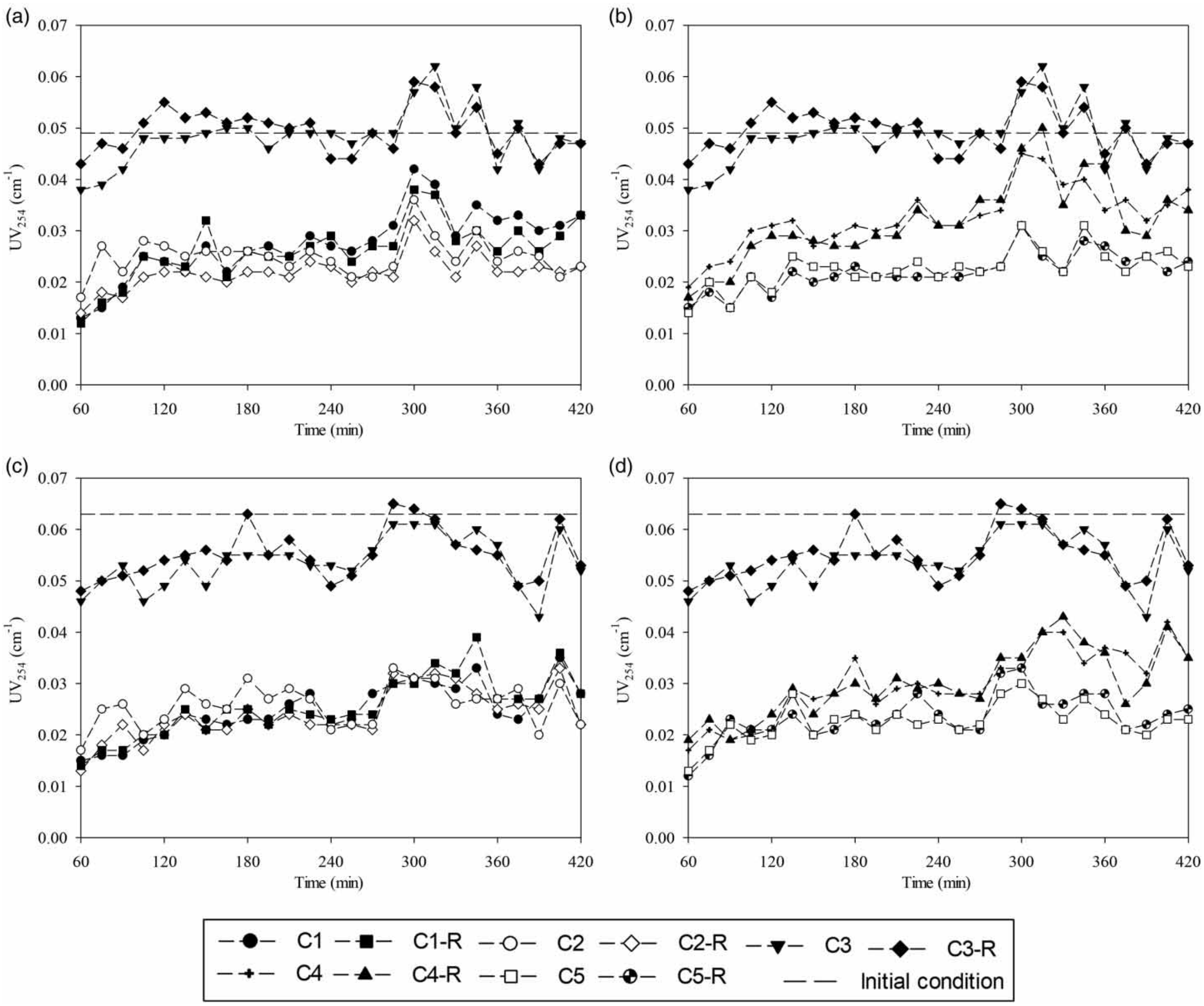

Figure 3 | UV_{254} measurements over time for all configurations using phenol concentrations 1 and 2. 1. Phenol at 0.25 mg/L. 2. Phenol at 1.20 mg/L.

Table 4 | TOC (mg/L) results over time for all the configurations using phenol concentration 1

Time (min)	C1	C1-R	C2	C2-R	C3	C3-R	C4	C4-R	C5	C5-R
135	1.248	0.932	1.255	1.147	2.760	2.766	1.783	1.360	1.452	1.021
240	1.135	0.960	0.952	0.903	2.651	2.735	1.525	1.348	0.833	0.879
330	1.138	0.937	0.720	0.644	2.436	2.554	1.382	1.416	0.854	0.750
420	1.270	1.129	0.668	0.628	2.543	2.575	1.566	1.819	0.694	0.722

However, the sand did not guarantee the reduction of phenols, which confirms the low efficiency of traditional filtering media used in water treatment processes, especially when the phenol concentration is higher (Christopher 2012).

The results of the ANOVA and Tukey tests showed that there were statistically significant differences for phenols between the sand and GAC configurations, whereas for TOC, both the sand and C4 configurations differed from

Table 5 | TOC (mg/L) results over time for all the configurations using phenol concentration 2

Time (min)	C1	C1-R	C2	C2-R	C3	C3-R	C4	C4-R	C5	C5-R
135	0.958	0.867	1.497	1.177	3.325	3.423	0.907	1.231	1.225	1.034
240	0.964	1.048	0.898	0.848	3.352	3.251	1.079	1.270	0.833	0.776
330	0.903	0.980	0.689	0.724	3.437	3.360	1.204	1.377	0.662	0.655
420	0.998	1.055	0.684	0.594	3.287	3.349	1.346	1.420	0.611	0.615

Table 6 | Phenols (mg/L) results over time for all the configurations using phenol concentration 1

Time (min)	C1	C1-R	C2	C2-R	C3	C3-R	C4	C4-R	C5	C5-R
135	0.0079	0.0032	0.0338	0.0152	0.3102	0.3397	0.0174	0.0081	0.0415	0.0251
240	0.0964	0.0343	0.0765	0.0047	0.3125	0.3096	0.0623	0.0347	0.0014	0.0118
330	0.0089	0.0592	0.0246	0.0775	0.3989	0.3560	0.0573	0.0366	0.0200	0.0095
420	0.0257	0.0362	0.0051	0.0099	0.3183	0.3484	0.0361	0.0234	0.0099	0.0099

Table 7 | Phenols (mg/L) results over time for all the configurations using phenol concentration 2

Time (min)	C1	C1-R	C2	C2-R	C3	C3-R	C4	C4-R	C5	C5-R
135	0.0370	0.0370	0.0453	0.0590	1.1908	1.2146	0.0504	0.0682	0.0543	0.0503
240	0.0096	0.0219	0.0100	0.0216	1.1822	0.9736	0.0058	0.0171	0.0169	0.0027
330	0.0138	0.0210	0.0190	0.0223	1.2285	1.2094	0.0242	0.0256	0.0287	0.0401
420	0.0099	0.0009	0.0097	0.0036	1.4216	1.3520	0.0148	0.0132	0.0068	0.0088

the other configurations. The sand reached maximum efficiencies for the removal of TOC and phenols of 26% and 9%, respectively, according to the UV_{254} results, confirming that the use of conventional filtration media, such as sand, is not suitable for the reduction of organic compounds during the process of treating drinking water.

With the GAC configurations, maximum efficiencies for the removal of TOC and phenols of 81% and 98%, respectively, were achieved for concentration 1 and of 85% and 99%, respectively, using concentration 2. Although no significant differences were found between the media with GAC, the maximum efficiencies were obtained using the configurations C1, C2 and C5 for TOC and C2 and C5 for phenols, indicating that better results are obtained using MAC and the 100% GAC configurations.

Studies such as that reported by Gibert *et al.* (2013) show that the effectiveness of GAC decreases over time during filtration, indicating the need to define the operating conditions for each type of water and filtration medium. In addition, the time to completely fill GAC adsorbent media and the phenol reduction efficiencies could be related to the presence of other organic compounds such as NOM in water because there is direct competition for the adsorption sites, blocking the pores and reducing the adsorption capacity of GAC (Guo *et al.* 2007; Zadaka *et al.* 2009).

CONCLUSIONS

Sand and GAC present different characteristics, which influence the efficiency of particle retention. The dual filters (sand and GAC) ensure better removal of material associated with dissolved organic matter (UV_{254} absorbance, TOC and phenols). With the GAC filters, maximum efficiencies of 93% regarding the UV_{254} values, 80% for TOC and 99% for phenols were achieved, while with sand, the

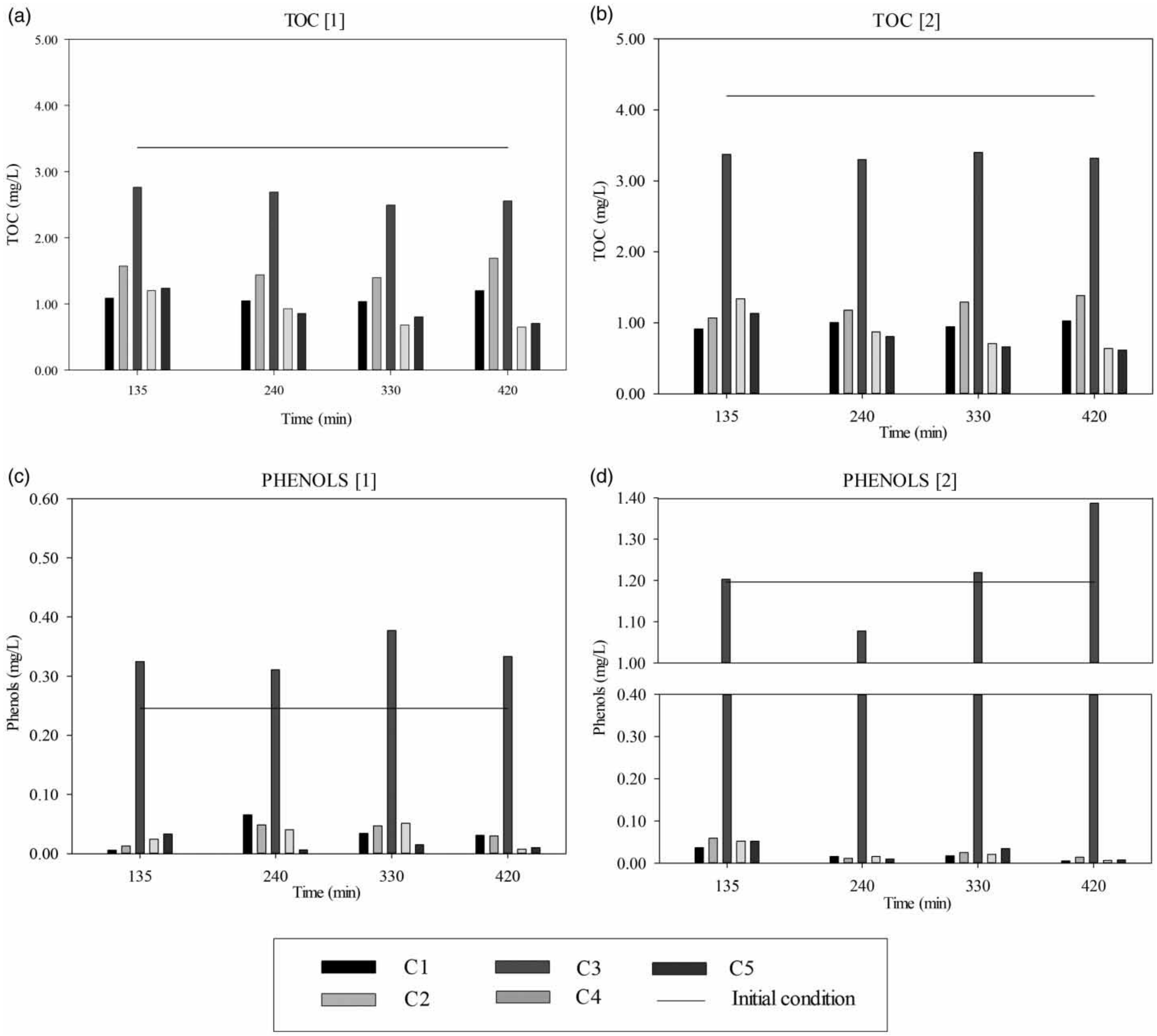

Figure 4 | Reduction of TOC and phenols over time for all configurations using phenol concentrations 1 and 2.

maximum efficiencies were 41% regarding the UV_{254} values, 26% for TOC and 10% for phenols. Considering the usefulness of the parameters indicating organic matter, correlation between such parameters is recommended because it is easy and fast to measure parameters such as UV_{254}, which is indicative of TOC, phenols and other organic compounds.

In general, it was noted that mineral GAC (TOC removal efficiency: 60–80%) was more efficient than vegetal GAC (TOC removal efficiency: 50–70%) independent of the configuration. Due to the adsorption efficiency depending on factors such as the contact time and the volume of the adsorbent, the operating conditions must be evaluated in each case to reach the limit values established by regulations.

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