Treatability of South African surface waters by activated carbon

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ABSTRACT

Natural organic matter (NOM) in water resources for drinking purposes can be removed by different methods, including activated carbon adsorption. Due to the variability of NOM in natural waters, both in terms of its nature and its concentration, a study was undertaken to investigate NOM removal for a wide range of South African surface waters, sampled at different periods, by the use of granular activated carbon (GAC). NOM removal was assessed by measuring the ultraviolet (UV) absorbance at 3 wavelengths, namely, 254 nm (UV_{254}), 272 nm (UV_{272}) and 300 nm (UV_{300}). A comparison of data between the three wavelengths showed that any of the three wavelengths can be used to assess NOM removal by GAC, which is well described by the Freundlich equilibrium equation. A treatment target of 40% removal of initial UV_{254} absorbance was considered. It was observed that, although the GAC dosage was generally a function of the initial UV_{254} absorbance, differences existed between waters. This suggests that GAC usage rate is not only a function of the initial UV absorbance but also of the NOM composition, indicating a need for improved NOM characterisation. Comparison between the UV absorbance and dissolved organic carbon (DOC) data suggested that for some waters UV_{254} absorbance can be used as a rapid substitute for DOC. Finally, the high GAC dosage rates required for the target criterion revealed that the process is inadequate for use at the initial stage of raw water treatment; GAC adsorption should be used at later stages of drinking water treatment.

Keywords: activated carbon, adsorption, Freundlich isotherm, natural organic matter, surface water, ultraviolet absorbance

INTRODUCTION

Natural organic matter is a complex mixture of organic compounds such as humic and fulvic acids, proteins, amino acids and carbohydrates, resulting from the degradation of plants, animals and microorganisms (Cornelissen et al., 2008; Edzwald and Tobiason, 2010; García et al., 2011). Based on its origin, NOM can be placed in two categories, namely autochthonous organic matter (formed within the water body) and allochthonous organic matter (produced elsewhere and transported to the water body) (Edzwald and Tobiason, 2010). It can be further classified as either particulate organic matter (POM) or dissolved organic matter (DOM). NOM in water affects the organoleptic aspects of the drinking water, promotes bacterial regrowth in drinking water distribution systems and reacts with disinfectants and oxidants, producing disinfection by-products and other products (Van der Kooij, 1998; Batterman et al., 2000; Hallam et al., 2001; Melnick et al., 2007; Edzwald and Tobiason, 2010; Ødegaard et al., 2010). NOM in raw water can be removed by different methods including the use of activated carbon. Adsorption of DOM by the activated carbon is a function of the NOM composition (nature and concentration), pH of the water, water temperature, molecular size and concentrations of some ions, such as magnesium and calcium (Schreiber et al., 2005).

In drinking water treatment plants, adsorption by activated carbon is a well-established process. The activated carbon is used either as powder (powdered activated carbon – PAC) or as granules (granular activated carbon – GAC). The PAC is generally used at the beginning of the treatment process (at or just after coagulation), while the GAC is used at a later stage of the treatment (generally before disinfection) (Kristiana et al. 2011; Matilainen et al., 2006). PAC is added to the water as slurry, while the GAC is placed in filter beds.

The aim of this study was to investigate NOM removal for a large range of South African surface waters by the use of GAC. The removal was assessed by measuring the UV absorbance at 254 nm (UV_{254}), 272 nm (UV_{272}) and 300 nm (UV_{300}). For some waters, the DOC values were used and compared with the UV absorbance values.

MATERIALS AND METHODS

Source water

Eight surface waters were sampled. As shown in Fig. 1, the sampling sites were chosen from different geographic regions in South Africa in order to include differences in NOM composition. The surface waters were also chosen to account for the main surface water types of South Africa (Oberholster, 2010). The different categories of waters are summarised in Table 1. The raw waters were collected at 5 different times to capture the seasonal variations in NOM composition (Sharp et al., 2006; Uyak et al., 2008). The sampling was done during the following periods: Round 1 from February to April 2010; Rounds 2, 3 and 4 in July 2010, November 2010 and February 2011, respectively. Round 5 waters were sampled in May and June 2011. Raw waters were collected (using two 25 ℓ plastic containers) and stored at approximately 4°C, in the dark. Analysis of the samples was done within 2 months, during which there was no significant change in NOM concentration or composition (Haarhoff et al., 2013).

Granular activated carbon preparation

The activated carbon used was a product commercially available in South Africa and kindly provided by a local supplier.

This paper was originally presented at the 2012 Water Institute of Southern Africa (WISA) Biennial Conference, Cape Town, 6–10 May 2012.

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http://dx.doi.org/10.4314/wsa.v39i3.6
Available on website http://www.wrc.org.za
ISSN 0378-4738 (Print) = Water SA Vol. 39 No 3 WISA 2012 Special Edition 2013
ISSN 1816-7950 (On-line) = Water SA Vol. 39 No 3 WISA 2012 Special Edition 2013

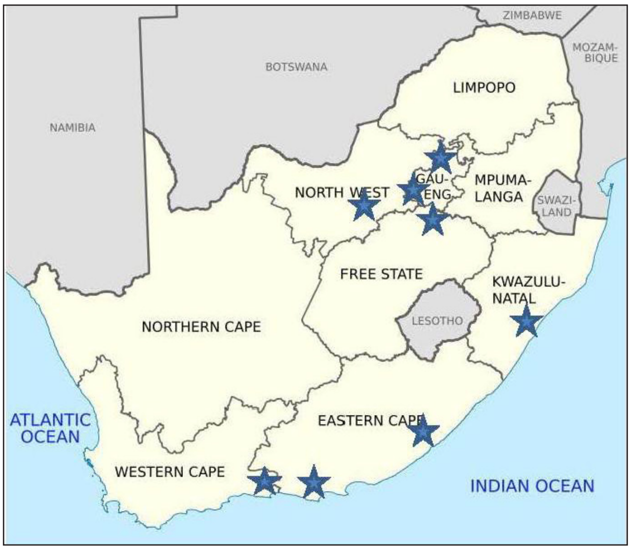

Figure 1
Sampling sites

(a)

(b)

Figure 2
Granular activated carbon, (a) before and (b) after grinding and sieving

It is a bituminous, coal-based GAC used in liquid phase applications to remove organic compounds. Figure 2 shows the GAC as supplied (a) and after grinding (b).

The general practice adopted for activated carbon studies is to grind the virgin GAC and use a fraction of the finely ground GAC. The main advantages of using the fine GAC are that it requires a shorter time to reach equilibrium with a smaller chance of biological interference. The unground GAC requires a longer time to reach equilibrium, which allows biological activity to take place and interfere significantly with the adsorption process (Randtke and Snoeyink, 1983). These authors found that the adsorptive capacity of the activated carbon was not significantly affected by grinding to a smaller size. When GAC is used in a column test, it is washed prior to use in order to remove the fine particles that can contaminate the effluent or plug the underdrain system. However, if the GAC is used in a batch equilibrium test, it is not necessary to apply this rinsing step since the fine particles are a very small fraction of the total mass of the carbon used.

To obtain the powdered activated carbon, 100 g of virgin GAC was crushed with a porcelain mortar and pestle. Using a vibratory sieve shaker, the fraction passing the 300 µm sieve was collected and stored in a glass container. The procedure was repeated until the quantity of fine GAC obtained was sufficient for further testing.

Batch adsorption tests

Batch tests were conducted at room temperature (20–25°C). On the day of testing, at least 3 ℓ of raw water was removed from refrigeration and stirred in the laboratory at 300 $r{\cdot}min^{-1}$ for a minimum of 30 min. Different masses of carbon (blank, 6.25, 8.25, 12.50, 16.50, 25.00, 33.25 and 50.00 mg) were each added to 250 mℓ of raw water in 500 mℓ Erlenmeyer flasks. The samples were placed on an orbital shaker table at 140 $r{\cdot}min^{-1}$ for 72 h (3 days). The rotation speed of 140 $r{\cdot}min^{-1}$ was chosen as it was visually observed to provide good mixing. After 3 days, the samples were filtered through a 0.45 µm hydrophilic Durapore (PVDF) membrane filter (Millipore Millex-HV) before the ultraviolet absorbance was measured at 254 nm (UV_{254}), 272 nm (UV_{272}) and 300 nm (UV_{300}) using a spectrophotometer (Ultrospec II UV/Vis). The DOC values of some samples were also measured using a total organic carbon analyser (Teledyne Tekmar, TOC fusion).

TABLE 1
Surface water categories

Plant names	Categories
Loerie (Water Treatment Plant)	Low-alkalinity low-colour water
Olifantsvlei (Wastewater Treatment Plant)*	Sewage effluent
Plettenberg Bay (Water Treatment Plant)	Very soft, highly coloured water
Rietvlei (Water Treatment Plant)	Eutrophic reservoir water
Stilfontein (Water Treatment Plant)	Eutrophic river water
Umzoniana (Water Treatment Plant)	Moderate-alkalinity low-colour
Vereeniging (Water Treatment Plant)	Oligotrophic reservoir water
Wiggins (Water Treatment Plant)	Low-alkalinity reservoir water

**The Olifantsvlei Wastewater Treatment Plant is not a direct drinking water source, but discharges into the Klip River and is therefore a worst-case indicator for rivers and streams which receive large volumes of treated sewage effluents.*

http://dx.doi.org/10.4314/wsa.v39i3.6
Available on website http://www.wrc.org.za
ISSN 0378-4738 (Print) = Water SA Vol. 39 No 3 WISA 2012 Special Edition 2013
ISSN 1816-7950 (On-line) = Water SA Vol. 39 No 3 WISA 2012 Special Edition 2013

The Freundlich equation was used for modelling the UV_{254} absorbance and shown in Eq. (1):

$$q_e = KC_e^n \quad (1)$$

where:

q_e = equilibrium NOM concentration in the solid phase (ℓ·mg^{-1}·m^{-1})
C_e = equilibrium NOM concentration in water (m^{-1})
K and n = Freundlich constant and exponent, respectively.

The Freundlich parameters K and n were determined from the results of absorbance at the three wavelengths. The parameters K and n are related to the capacity and affinity of the carbon for NOM molecules, respectively (Cornelissen et al., 2008).

Performance indicator

The required GAC dosage for each raw water was determined for an arbitrary treatment goal of 40% removal of initial UV_{254} absorbance.

The GAC dosage calculation was derived from Eq. (1) as follows:

$$\frac{C_i - C_e}{M} = KC_e^n \quad (2)$$

$$M = \frac{C_i - C_e}{KC_e^n} \quad (3)$$

If C_i is removed by 40%

$$M = \frac{C_i - 0.60C_i}{K(0.60C_i)^n} \quad (4)$$

where:

C_i = initial NOM concentration in raw water (m^{-1})
M = GAC dosage (mg·ℓ$^{-1}$)

RESULTS AND DISCUSSION

Comparison of absorbance results at three wavelengths

The indicators used and reported in this paper are the UV_{254} absorbance (indicative of the presence of conjugated C=C double bonds or compounds with aromatic structure) (Edzwald and Tobiason, 2010), UV_{272} absorbance (reported by some as the best indicator of total organic halogen and trihalomethane formation (Korshin et al., 1997)) and UV_{300} absorbance (used by some South African water treatment plants as an operational parameter) (Haarhoff et al., 2012). Ultraviolet absorbance at 254 nm (UV_{254}) is used by many organisations to characterise NOM (Karanfil et al., 2002). It was found that, when plotting the graphs of equilibrium UV absorbance in water (C_e) versus the GAC dosage, the removal patterns were the same for all wavelengths, as shown in Figs. 3 and 4 (Round 5 Stilfontein and Round 3 Umzoniana waters, respectively).

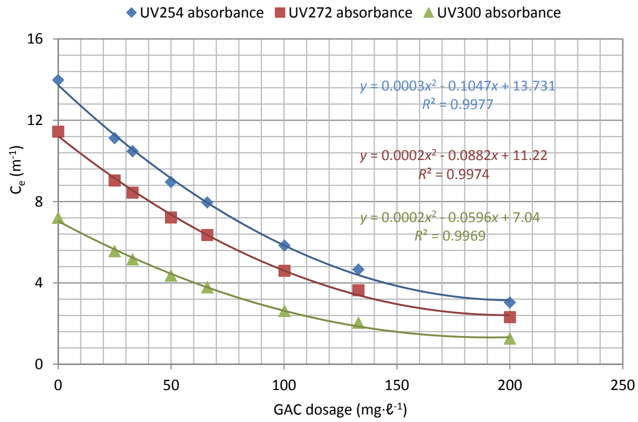

Figure 3
Equilibrium UV absorbance vs. GAC dosage for the Round 5 Stilfontein water

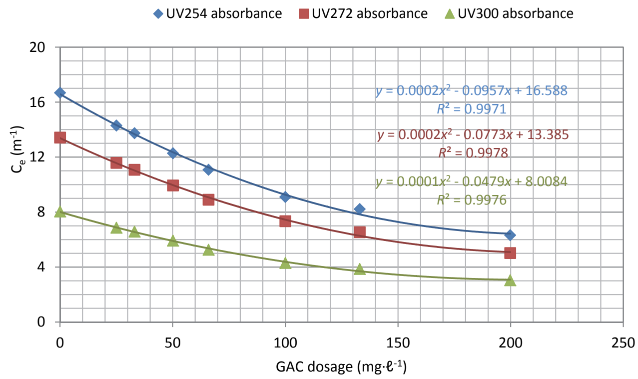

Figure 4
Equilibrium UV absorbance vs. GAC dosage for the Round 3 Umzoniana water

TABLE 2
Ratios between UV_{272} and UV_{254} and, UV_{300} and UV_{254} absorbance results

GAC dosage (mg·ℓ$^{-1}$)	Round 5 - Stilfontein water		Round 3 - Umzoniana water	
	UV_{272}/UV_{254}	UV_{300}/UV_{254}	UV_{272}/UV_{254}	UV_{300}/UV_{254}
0	0.82	0.52	0.80	0.48
25	0.81	0.50	0.81	0.48
33	0.81	0.49	0.81	0.48
50	0.81	0.49	0.81	0.48
66	0.80	0.47	0.80	0.47
100	0.79	0.45	0.81	0.47
133	0.78	0.44	0.80	0.47
200	0.76	0.41	0.79	0.48

Table 2 gives the ratios between UV_{272} and UV_{254} and, UV_{300} and UV_{254} for the above-mentioned waters. It demonstrates that the ratios between UV_{272} and UV_{254} were practically the same for all the GAC dosages for these two water samples. The same conclusion applied to the ratios between the UV_{300} and UV_{254}. The average value, for all the water samples, of the ratio between UV_{272} and UV_{254} was 0.82 (SD = 0.0006), and for the ratio between UV_{300} and UV_{254} was 0.53 (SD = 0.0033).

Table 3 presents the UV absorbance removal at different wavelengths for certain waters. UV absorbance removal was the same for a particular water at the same GAC dosage for the three wavelengths.

Table 4 compares the GAC dosages required to remove 40% of the initial UV absorbance at 3 wavelengths for 3 different waters. The dosages were calculated by applying the Freundlich equation.

Table 4 indicates that the dosage required to remove the initial UV absorbance by a fixed percentage (in this case 40%) was approximately the same for all three wavelengths for each water sample. It also shows that the required dosage varied

TABLE 3
UV absorbance removal (%)

GAC dosage (mg·ℓ⁻¹)	Initial removal (%)								
	Round 1 – Loerie			Round 2 – Rietvlei			Round 4 – Olifantsvlei		
	UV_{254}	UV_{272}	UV_{300}	UV_{254}	UV_{272}	UV_{300}	UV_{254}	UV_{272}	UV_{300}
25	13	13	12	-	-	-	-	-	-
33	17	17	17	23	24	24	25	26	25
50	19	19	18	30	30	31	31	32	31
66	26	27	26	39	40	40	40	40	39
100	36	35	34	50	51	51	51	52	50
133	44	43	41	60	60	60	57	58	55
200	58	58	55	71	72	71	67	67	64

TABLE 4
Dosages required to remove 40% of the initial UV absorbance

UV absorbance	Round 5 Wiggins	Round 5 Olifantsvlei	Round 2 Plettenberg Bay
	M (mg·ℓ⁻¹)	M (mg·ℓ⁻¹)	M (mg·ℓ⁻¹)
UV_{254}	30	58	189
UV_{272}	28	54	194
UV_{300}	26	50	212
Average	28	55	198

TABLE 5
Freundlich parameters derived from UV_{254} absorbance data‡

Plant name	Round 1		Round 2		Round 3		Round 4		Round 5	
	K	N	K	n	K	n	K	n	K	n
Umzoniana	1.2E-2	0.86	1.9E-2	0.64	1.5E-2	0.71	9.4E-3	0.88	2.3E-3	1.38
Wiggins	1.4E-2	1.03	2.2E-2	0.95	1.2E-2	1.22	5.0E-3	1.61	4.1E-2	0.43
Loerie	1.2E-2	0.75	1.2E-2	0.88	1.0E-2	0.83	1.5E-2	0.71	3.5E-2	0.32
Rietvlei	2.8E-2	0.51	2.2E-2	0.63	-	-	2.2E-2	0.57	-	-
Vereeniging	3.4E-2	0.41	-	-	1.5E-2	1.71	-	-	1.2E-2	0.85
Olifantsvlei	2.5E-2	0.73	1.8E-2	0.79	1.6E-2	0.78	1.1E-2	0.97	2.4E-2	0.65
Stilfontein	2.8E-2	0.58	1.5E-2	0.80	1.4E-2	0.82	3.4E-2	0.46	3.0E-2	0.55
Plettenberg Bay	-	-	7.2E-3	0.75	-	-	-	-	2.4E-5	2.23

‡Cells with no data had R^2 values that were very poor ($R^2 \leq 0.11$) or there was no data. The non-shaded cells represent those samples where the R^2 values were very good (R^2 range 0.91–1.00). The shaded cells were those samples where R^2 was less than 0.90.

TABLE 6
GAC dosage *M* (mg·ℓ⁻¹) for removal of 40% of UV_{254} absorbance

Plant name	Round 1		Round 2		Round 3		Round 4		Round 5	
	C_i	M	C_i	M	C_i	M	C_i	M	C_i	M
Umzoniana	15.8	78	14.8	75	16.7	89	19	96	28.2	-
Wiggins	7.0	46	6.3	33	6.1	42	7.6	53	4.9	30
Loerie	25.2	-	17.3	71	6.7	82	15.1	83	13.3	79
Rietvlei	18.4	79	17.2	70	19.2	-	24.0	94	19.7	-
Olifantsvlei	13.1	48	16.4	61	15.8	68	15.4	67	14.3	58
Stilfontein	16.4	63	15.3	68	18.5	72	33.7	-	14.0	58
Plettenberg Bay	-	-	30.4	189	43.8	-	62.9	-	49.3	-

with the water type. To remove 40% of initial UV absorbance, Round 5 Olifantsvlei water required almost twice the dosage of the Wiggins Round 5 water, whilst the Plettenberg Bay Round 2 needed approximately 7 times more GAC than the Round 5 Wiggins water. These differences of dosages show that NOM composition varies with water source. Thus, improved NOM characterisation is required.

UV_{254} absorbance values

Based on all the findings presented above, it appeared that any of the three wavelengths investigated could be used to assess NOM removal. In this paper, the rest of the data analysis was done with the UV_{254} absorbance value, as this is the most popular wavelength used in the published literature (Karanfil et al., 2002). The Freundlich equilibrium equation was then applied to UV_{254} absorbance data. Table 5 presents the Freundlich parameters of the water samples. Round 1 Loerie and Vereeniging had an R^2 of 0.80 and 0.73, respectively, while Round 3 of the Vereeniging water had an R^2 of 0.78. The lowest R^2, 0.61, for Round 4 was found for the Stilfontein water. For Round 5, the Umzoniana, Vereeniging and Plettenberg Bay waters displayed R^2 values of 0.78, 0.83 and 0.66, respectively.

Performance criterion

The required GAC dosage was calculated, i.e. the carbon dosage required to remove the initial UV_{254} absorbance (C_i) by 40%. Table 6 represents the calculated GAC dosage *M* (mg·ℓ⁻¹) to meet the required criterion.

In general, the smaller the initial UV_{254} absorbance, the smaller the required GAC dosage, e.g. all of the Wiggins water samples. When the UV absorbance increased the dosage also increased. There were, however, some notable exceptions: Round 1 Olifantsvlei water with initial UV_{254} absorbance of 13.1 m⁻¹ required a lower dosage of 48 mg·ℓ⁻¹ and Umzoniana Round 2 with initial UV_{254} absorbance of 14.8 m⁻¹ required a higher dose

http://dx.doi.org/10.4314/wsa.v39i3.6
Available on website http://www.wrc.org.za
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ISSN 1816-7950 (On-line) = Water SA Vol. 39 No 3 WISA 2012 Special Edition 2013

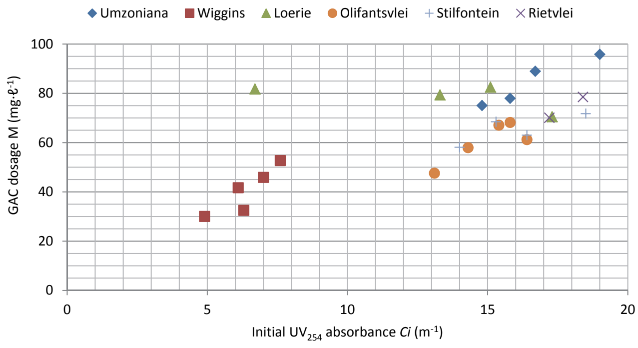

Figure 5
GAC dosage vs. initial UV_{254} absorbance

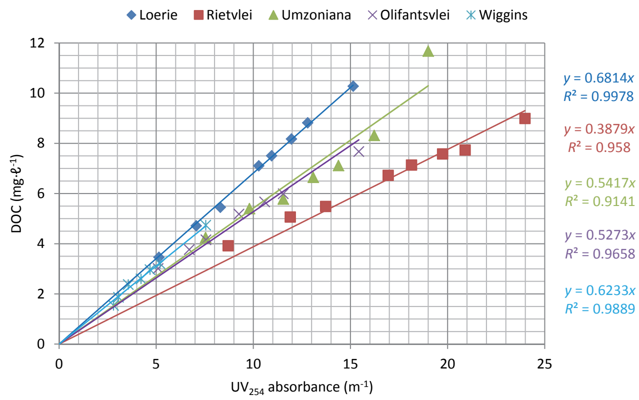

Figure 6
Relationship between DOC and UV_{254} absorbance for some Round 4 waters

of 75 mg·ℓ⁻¹. The GAC usage of all of the samples with $R^2 < 0.90$ is not shown in Table 6.

Figure 5 presents initial UV_{254} absorbance versus GAC dosage. Some waters displayed a clear relationship between the activated carbon dosage and the initial UV_{254} absorbance. It was, therefore, possible to predict the GAC usage rate of some waters, but for others it was difficult. The data of the Plettenberg Bay water Round 2 (initial UV_{254} absorbance and GAC usage of 30.4 m⁻¹ and 189.4 mg·ℓ⁻¹, respectively) was not included in Fig. 6 because it was out of the range of the figure.

However, it appears that the dosages required for achieving the target criterion are high compared to what can be tolerated by water treatment plants.

Comparison of DOC and UV_{254} absorbance

Parallel DOC analysis was performed on the Round 4 waters (i.e. Loerie, Rietvlei, Umzoniana, Olifantsvlei and Wiggins). Figure 6, representing the comparison between the DOC and UV_{254} absorbance data of the selected waters, showed that there was a strong linear relationship between the two surrogate parameters, for NOM characterisation.

It was also found that the GAC dosage required to remove 40% of UV_{254} absorbance also removed about 40% of the DOC (see Table 7). This suggests that UV_{254} absorbance might be used as a quick and reliable characterisation parameter for some South African waters. The specific UV absorbance (SUVA) values of the waters presented in the last column of Table 7 are good indicators of the humic acid content in water (USEPA, 1999). The SUVA values suggested that there were two categories of waters: those with a high fraction of non-humic matter (SUVA < 2) and those with a mixture of aquatic humic and non-humic matter (SUVA between 2 and 4).

TABLE 7
DOC and UV_{254} absorbance removal

Plant name	Initial UV_{254} (m⁻¹)	Dosage for 40% UV_{254} removal (mg·ℓ⁻¹)	Initial DOC (mg·ℓ⁻¹)	DOC removed	SUVA (ℓ·mg⁻¹m⁻¹)
Wiggins	7.6	53	4.74	40%	1.60
Umzoniana	19.0	96	11.68	50%	1.63
Loerie	15.1	83	10.28	39%	1.47
Rietvlei	24.0	94	8.99	36%	2.67
Olifantsvlei	15.4	67	7.68	34%	2.01

CONCLUSION

The aim of the study was to investigate the removal of NOM by the use of GAC by measuring the UV absorbance at 254 nm (UV_{254}), 272 nm (UV_{272}) and 300 nm (UV_{300}), for a representative selection of South African surface waters, in batch experiments. The performance of activated carbon was considered over a range of dosages in order to detect some general patterns – not to suggest practical or economical values. The data for the three wavelengths were compared first:

- It was found that, at all three wavelengths, the percentage of NOM removal was practically the same. The differences amongst these wavelengths can, therefore, not be used to characterise the differences in NOM composition of the different sources. Any of the three wavelengths can be used to assess the NOM removal by GAC.
- The UV_{272} and UV_{300} absorbance values were, on average, 0.82 and 0.53 times the UV_{254} absorbance data, respectively. The UV_{254} absorbance data were chosen for the remainder of the analysis, as it is the most popular wavelength used in the published literature.

The Freundlich equilibrium equation provided a good mathematical description of the adsorption. The granular activated carbon dosage could be calculated for an arbitrary goal of 40% of UV absorbance removal by fitting this isotherm to the UV_{254} absorbance data. It was found that:

- The value of the initial UV absorbance value impacts on the dosage rate. Results suggested that when the initial UV absorbance is low, the GAC dosage required is also likely to be small.
- The GAC usage rate is not only dependent on the initial UV_{254} absorbance of the water but also on the composition (nature and concentration) of the NOM indicating a need for improved NOM characterisation.

Dissolved organic carbon analysis was conducted on some waters:

- Strong linear relations were found between the DOC and the UV_{254} absorbance values, suggesting that, for those waters, the latter could be used as a quick and reliable characterisation surrogate parameter for NOM removal.
- The dosages required to remove 40% of UV absorbance also removed approximately the same percentage of DOC.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the financial support from the University of Johannesburg and the South African Water Research Commission.

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http://dx.doi.org/10.4314/wsa.v39i3.6
Available on website http://www.wrc.org.za
ISSN 0378-4738 (Print) = Water SA Vol. 39 No 3 WISA 2012 Special Edition 2013
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