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Published PDF deposited in Coventry University's Repository

Original citation:

Salvestrini, S, Vanore, P, Bogush, A, Mayadevi, S & Campos, LC 2017, 'Sorption of metaldehyde using granular activated carbon', Journal of Water Reuse and Desalination, vol. 7, no. 3, pp. 280-287.

<https://dx.doi.org/10.2166/wrd.2016.074>

DOI 10.2166/wrd.2016.074

ISSN 2220-1319

ESSN 2408-9370

Publisher: IWA Publishing

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Sorption of metaldehyde using granular activated carbon

S. Salvestrini, P. Vanore, A. Bogush, S. Mayadevi and L. C. Campos

ABSTRACT

In this work, the ability of granular activated carbon (GAC) to sorb metaldehyde was evaluated. The kinetic data could be described by an intra-particle diffusion model, which indicated that the porosity of the sorbent strongly influenced the rate of sorption. The analysis of the equilibrium sorption data revealed that ionic strength and temperature did not play any significant role in the metaldehyde uptake. The sorption isotherms were successfully predicted by the Freundlich model. The GAC used in this paper exhibited a higher affinity and sorption capacity for metaldehyde with respect to other GACs studied in previous works, probably as a result of its higher specific surface area and high point of zero charge.

Key words | diffusion-controlled sorption, environmental pollution, granular activated carbon, metaldehyde, sorption

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INTRODUCTION

Metaldehyde is a cyclic tetramer of acetaldehyde, commonly used as molluscicide in agriculture and domestic gardening to control slugs, snails and other gastropods. Because it is an environmental contaminant, its current maximum application rate is fixed to 700 g metaldehyde/ha/calendar year in the UK (The Metaldehyde Stewardship Group 2013).

Since 2008, the UK Environmental Agency has been reporting that the metaldehyde level in drinking water exceeds European and UK limits of $0.1 \mu\text{g L}^{-1}$ (Busquets *et al.* 2014). This contaminant is inefficiently treated in water treatment plants. In fact, the maximum reported concentration of metaldehyde in UK water treated for drinking use is just above $1.03 \mu\text{g L}^{-1}$ (Busquets *et al.* 2014).

There are different methods designed for reducing the concentration of metaldehyde in water (Autin *et al.* 2012;

Tao & Fletcher 2013, 2014; Busquets *et al.* 2014). An important method to reduce the concentration of metaldehyde is photocatalytic degradation using UV/TiO₂ or UV/H₂O₂ systems. Autin *et al.* (2012) showed that both UV/H₂O₂ and UV/TiO₂ are able to efficiently degrade metaldehyde in pure systems. However, in natural waters, the UV/TiO₂ process is severely inhibited and the use of UV/H₂O₂ remains the only reliable option for metaldehyde removal, although both these processes are relatively expensive compared to the conventional processes.

Among other techniques proposed for metaldehyde removal from water, sorption is the most interesting one because it is generally cheap and easy to scale up. Tao & Fletcher (2013) studied the sorption of metaldehyde onto three different materials: (1) granular activated carbon (GAC); (2) non-functionalised hyper-cross-linked polymer; (3) ion-exchange resin. Their results indicated that the sorption kinetics of metaldehyde onto all tested materials was fast, reaching equilibrium within 8 hours. They also

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doi: 10.2166/wrd.2016.074

showed that ion-exchange resin exhibited the highest sorption capacity ($1,807 \text{ mg g}^{-1}$) and GAC was relatively inefficient in removing metaldehyde (71 mg g^{-1}). In terms of operational costs, ion-exchange resins are generally slightly cheaper than GAC (Griffin 2009). However, on the other hand, GAC is less selective and hence more capable of adsorbing different types of pollutants.

The aim of this work was to investigate the sorption of metaldehyde onto a different commercial GAC by varying the amount of GAC, initial solute concentration, pH, ionic strength and temperature.

Table 1 | Main physicochemical characteristics of metaldehyde

Metaldehyde

Molecular formula	$\text{C}_8\text{H}_{16}\text{O}_4$
Molecular mass	176.21 g/mol
Water solubility (25°C)	213 mg L^{-1} ^a
Appearance	White or colourless crystalline solid
Toxicity	Moderately toxic, kidney and liver toxicant ^a

^aUniversity of Hertfordshire, Pesticide Properties Database-Metaldehyde, UK, 2012.

MATERIALS AND METHODS

Materials and reagents

Metaldehyde (CAS 9002-91-9) was supplied by Sigma-Aldrich; its main characteristics are reported in Table 1.

The cartridges, styrene-divinylbenzene (SDB1) used for the solid phase extraction (SPE) were provided by J. T. Baker (USA), while analytical (HPLC) grade methanol (CAS 67-56-1) and dichloromethane (CAS 75-09-2) were purchased from Fisher Scientific (UK).

The coal based GAC used in this study was obtained from Chemviron Carbon (UK) and was used as received. The GAC particle size ranged from 1 to 2 mm. The Brunauer–Emmett–Teller (BET) surface area of the GAC was $774 \text{ m}^2/\text{g}$. The GAC particle morphology with element analysis using scanning electron microscopy/energy dispersive X-ray spectroscopy (SEM/EDX) is presented in Figure 1. The GAC particles had a porous surface with a pore size $<200 \mu\text{m}$. The GAC mainly consisted of C with impurities of Ca, Al, Si, and S.

The determination of the pH of the point of zero charge (pHpzc) of adsorbents was carried out by pH titration procedures (Rivera-Utrilla *et al.* 2001). Forty mL aliquots of 0.01 M NaCl solution were placed in 50 mL conical tubes

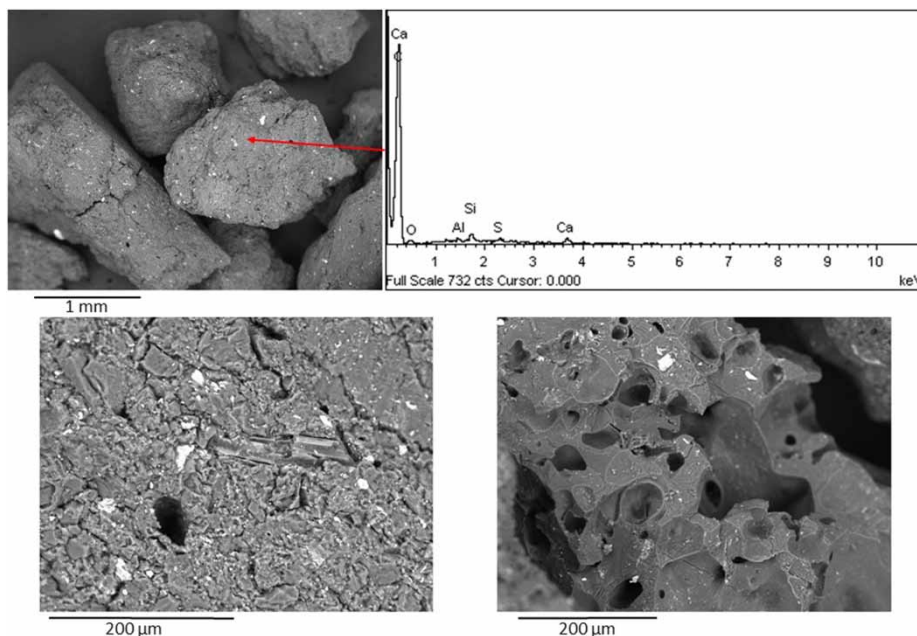


Figure 1 | Morphology and element analysis of the GAC particle.

and the pH was adjusted to a value between 2 and 10 by the addition of a few drops of 0.1 M HCl or 0.1 M NaOH solution. One hundred and twenty milligrams of GAC were added to each solution, and after 2 days the final pH was measured and plotted against the initial pH. The pH at which the curve pH final vs pH initial crosses the line pH final = pH initial represents the point of zero charge (PZC) of GAC.

Kinetics and sorption of metaldehyde

Metaldehyde sorption was studied by batch method. Kinetic experiments were performed by adding 10 mg of GAC to 100 mL of metaldehyde solution (30 mg L⁻¹). The samples were agitated in a shaker (30 rpm) at room temperature and, at pre-decided contact times, 0.2 mL aliquots of reacting solution were removed for gas chromatography–mass spectrometry (GC-MS) analysis.

Metaldehyde sorption at equilibrium was studied by contacting 1–10 mg of GAC with 10 mL of metaldehyde solution (1–30 mg L⁻¹). The samples were stirred at 30 rpm until the attainment of equilibrium, and analyzed as described above. Sorption was studied as a function of temperature (5, 25, 45 °C), pH (2–8) and ionic strength (0–1 M). The pH of samples was adjusted to the desired value with a few drops of concentrated HCl or NaOH. KCl was used to alter the ionic strength of the solution.

GC-MS analysis

Before GC-MS analysis, aliquots collected from the samples were pre-concentrated using SPE according to the following procedure. A styrene-divinylbenzene (SDB1) cartridge was first activated, flushing 10 mL of methanol and then conditioned with 2 mL of Mill-Q water. Afterwards, 0.2 mL of sample was passed through the cartridge. The cartridge was then rinsed with 2 mL of Mill-Q water (to ensure that metaldehyde was entirely retained on the polymer) and dried by passing air through it for 40 min. Finally, the cartridge was flushed with 3 mL of dichloromethane; the fraction was collected in an appropriate glass tube and evaporated to 1 mL by nitrogen for GC-MS analysis. The GC-MS equipment used was a Perkin Elmer Clarus 500 which included an auto-injector, mass-spectrometer capable, a selective ion monitoring mode, and a column HP5-MS (30 m × 0.25 mm diameter, 0.25 μm film thickness).

The injection model was split-less and the temperatures of injector and detector were set at 100 and 180 °C, respectively. Helium was used as the carrier gas (1 mL min⁻¹). The temperature programme for the oven was set at 100 °C and held for 1 min; then increased to 150 °C at a rate of 5 °C min⁻¹ and held for 1 min.

RESULTS AND DISCUSSION

Sorption kinetics

A first attempt to model the sorption kinetic data was carried out using a pseudo-first and pseudo-second order equations.

According to the pseudo-first model (Lagergren 1898), the sorption rate is proportional to the difference of the amount sorbed at equilibrium (q_e , μg g⁻¹) and at time t (q_t , μg g⁻¹):

$$\frac{dq_t}{dt} = k_1(q_e - q_t) \quad (1)$$

where k_1 (h⁻¹) represents the pseudo-first kinetic rate constant.

Integrating Equation (1) with the boundary conditions $q_t = 0$ at $t = 0$ and $q_t = t$ at $t = t$ leads to:

$$q_t = q_e(1 - e^{-k_1 t}) \quad (2)$$

The pseudo-second order kinetics (Ho & McKay 1998) in its differential form is given by:

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2 \quad (3)$$

It can be integrated to give:

$$q_t = \frac{k_2 t q_e^2}{1 + k_2 t q_e} \quad (4)$$

where k_2 (μg g⁻¹ h⁻¹) is the pseudo-second order kinetic rate constant.

Figure 2(a) and 2(b) show the experimental kinetic data modelled according to the pseudo-first and pseudo-second equation, respectively. The equilibrium was achieved within about 48 h. The experimental data were better described by the pseudo-second order equation, as inferred by the higher correlation coefficient R^2 and by the lower parameter errors (see Table 2).

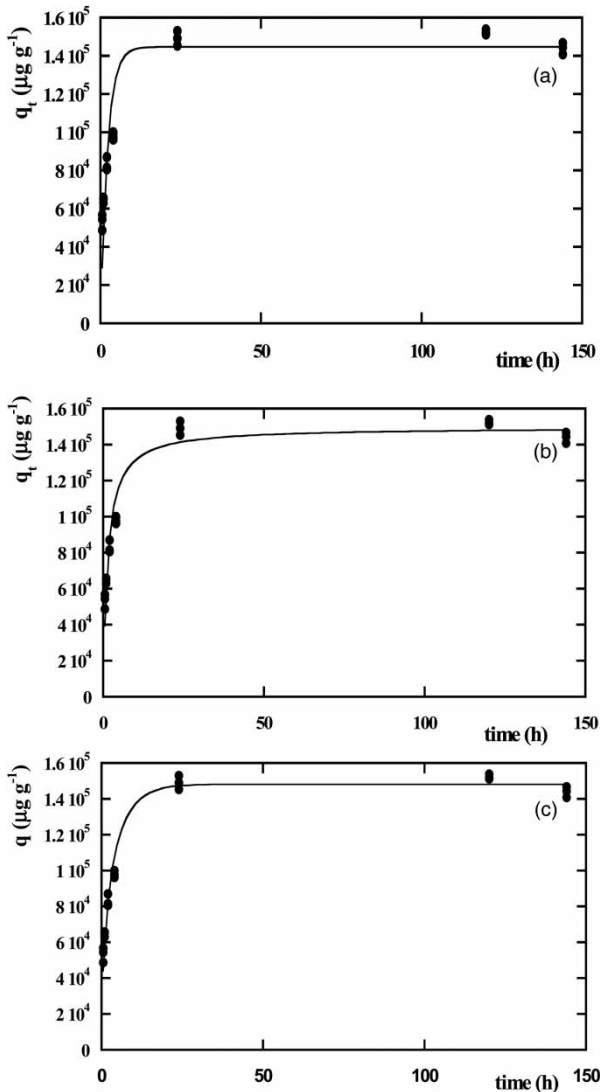


Figure 2 | (a) Pseudo-first order, (b) pseudo-second order and (c) Vermeulen kinetic models for metaldehyde sorption.

In order to confirm that the pseudo-second order equation was appropriate for describing the experimental data, the linearized form of this model as a control tool was examined.

The most used linearized form of Equation (4) is as follows (Vidal *et al.* 2011; Mobasherpour *et al.* 2014):

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (5)$$

If the data obey the pseudo-second order model, a plot of t/q vs t should produce a straight line. This is consistent with the results reported in Figure 3(a).

However, it has been demonstrated (Canzano *et al.* 2012) that the use of Equation (5) may lead to incorrect conclusions, especially when the sorption data are at equilibrium (or very close to) because in such cases the plot t/q vs t becomes linear irrespective of the sorption kinetics.

A better option for reporting the experimental data may be to use the following linearized form:

$$\frac{q}{t} = k_2 q_e^2 - k_2 q_e q_t \quad (6)$$

The results using this model are presented in Figure 3(b).

It can be seen from Figure 3(b) that the data significantly deviate from linearity, especially at higher sorption amounts. This fact clearly suggests that the pseudo-second order model does not exhaustively describe the experimental data trend. This could be explained by the fact that the rate of sorption of metaldehyde onto GAC may be controlled by diffusion. In support of this hypothesis, it should be noted that GAC has a microporous structure (Figure 1) which could offer a resistance (via intra-particle diffusion) to the mass transfer of metaldehyde and, hence, limit the overall reaction rate. Under this hypothesis, and assuming that the sorbent particles are uniform and spherical and that the solute uptake is small relative to the total quantity introduced into the system, the sorption rate can be described by (Ruthven 1984):

$$\frac{q_t}{q_\infty} = 1 - \frac{6}{\pi^2} \sum \exp\left(-\frac{D_c \pi^2 t}{r_c^2}\right) \quad (7)$$

where D_c ($\text{m}^2 \text{h}^{-1}$) and r_c (m) are the intracrystalline diffusivity and mean radius of the sorbent particle, respectively.

Table 2 | Kinetic parameters for metaldehyde sorption as determined by the fitting procedure

Model	q_e ($\mu\text{g g}^{-1}$)	k_1 (h^{-1})	k_2 ($\text{g } \mu\text{g}^{-1} \text{h}^{-1}$)	Z (h^{-1})	k_D ($\mu\text{g g}^{-1} \text{h}^{-0.5}$)	I ($\mu\text{g g}^{-1}$)	R^2
Pseudo-first order model	$(1.45 \pm 0.05) \times 10^5$	0.45 ± 0.05					0.870
Pseudo-second order model	$(1.49 \pm 0.03) \times 10^5$		$(4.8 \pm 0.5) \times 10^{-6}$				0.950
Vermeulen model	$(1.48 \pm 0.02) \times 10^5$			0.18 ± 0.01			0.975
Weber–Morris model					$(3.5 \pm 0.2) \times 10^4$	$(3.0 \pm 0.3) \times 10^4$	0.965

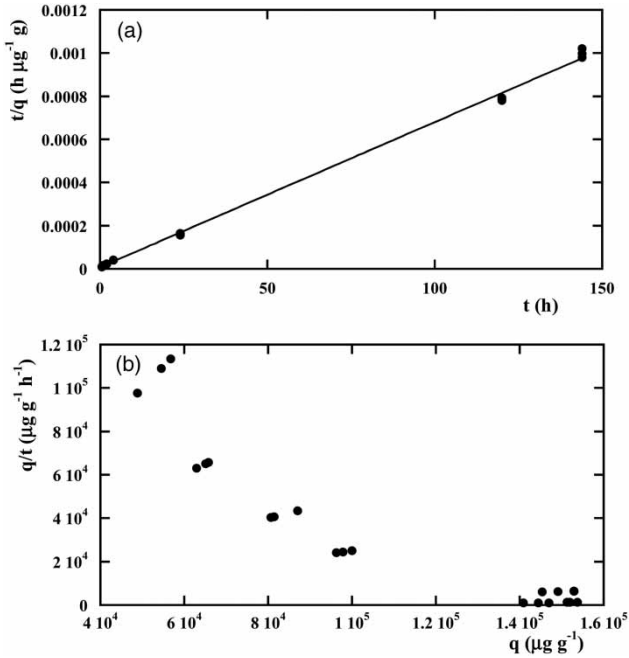


Figure 3 | (a) and (b) Plots of metaldehyde kinetic sorption data: a) t/q vs t plot; b) q/t vs t plot.

If the q_t/q_e ratio is >0.1 , the Vermeulen approximation can be applied (Poulopoulos & Inglezakis 2006) so that Equation (7) reduces to:

$$q_t = q_e \sqrt{1 - \exp(-Zt)} \quad (8)$$

where Z (h^{-1}) is $D_c \pi^2 / r_c^2$.

The above equation was used to model the experimental kinetic data and the results are reported in Figure 2(c) and Table 2. It can be seen that Equation (8) gives a better fit compared to the pseudo-first and pseudo-second order models both in terms of R^2 and parameter errors. This leads us to conclude that diffusion plays a major role in the sorption rate of metaldehyde onto GAC.

The results of our study indicate that the uptake of metaldehyde takes place slowly compared to that measured using a different commercial GAC (henceforth GAC-2; Tao & Fletcher 2013). For our GAC, the time required to reach equilibrium was about 24 h (Figure 2), whereas GAC-2 had a shorter (about 6 h) equilibration time. Having assumed that diffusion controls the rate of sorption, it is reasonable to ascribe the different behaviour of the two compared sorbents to their particle size as the rate of the process, in that case, should vary inversely with particle size (Boyd et al. 1947).

Consistent with the results of the kinetic experiments, the GAC used in the present work has greater particle size (1–2 mm) than that of GAC-2 (0.4–0.8 mm).

In order to strengthen the hypothesis that metaldehyde uptake is diffusion-controlled, initial sorption data (i.e. far from equilibrium) should be proportional to the square root of time according to the further simplification of Equation (7) derived by Weber–Morris (Weber & Morris 1963; Salvestrini et al. 2014):

$$q_e = k_D \sqrt{t} + I \quad (9)$$

where k_D ($\mu\text{g g}^{-1} \text{h}^{-0.5}$) is the kinetic intra-particle diffusion constant and I ($\mu\text{g g}^{-1}$) a constant proportional to the thickness of the boundary layer.

Therefore, a plot of q vs $t^{0.5}$ should yield a straight line with slope and intercept with the q axis equal to k_D and I , respectively. Figure 4 shows the Weber–Morris plot for the metaldehyde uptake onto GAC. The plot shows a good linearity in the range of time selected, thus confirming that, in the sorption process, intra-particle diffusion is the rate-limiting step.

Sorption isotherms

The equilibrium data of metaldehyde sorption onto GAC were modelled using Langmuir, Freundlich and Langmuir–Freundlich models (Equations (10)–(12), respectively):

$$q_e = \frac{q_{\max} k_L C_e}{1 + k_L C_e} \quad (10)$$

$$q_e = k_F C_e^N \quad (11)$$

$$q_e = \frac{q_{\max} K_{LF} C_e^N}{1 + K_{LF} C_e^N} \quad (12)$$

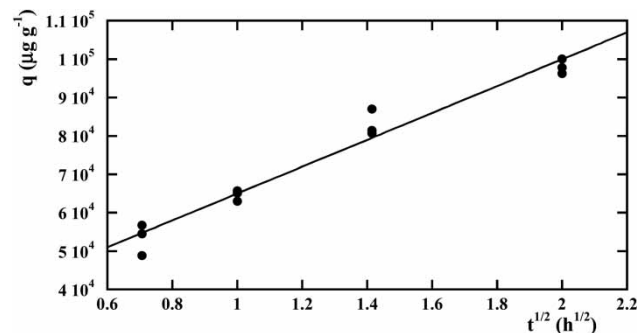


Figure 4 | Weber–Morris plot for metaldehyde sorption.

where q_e is the amount of the sorbate at the equilibrium, K_L ($L \mu g^{-1}$), K_F ($mg^{1-N} g^{-1} L^N$) and K_{LF} ($L^N \mu g^{-N}$) are the sorption affinity constants, q_{max} is the amount of sorbate necessary to complete a monolayer and N is a dimensionless constant related to the heterogeneity of the sorbent.

Figure 5(a) and 5(b) show the experimental data fitted with the Langmuir and Freundlich models, respectively. Metaldehyde sorption onto GAC was better described by the Freundlich model, especially at low solute concentration. These findings suggest the GAC behaves as heterogeneous material consisting of sorption sites which are not energetically equivalent (Toth 2002).

Having established that the Langmuir model is not suitable to represent the sorption data, the Langmuir-Freundlich model was used in order to gain information on the saturation level of the sorbent.

Based on the results of the fitting procedure (Figure 5(c) and Table 3), GAC has a q_{max} value of 320 mg g^{-1} . It is worth noting that the value of q_{max} calculated in this work is much higher than the value of 71 mg g^{-1} (Tao & Fletcher 2013) and 15 mg g^{-1} (Busquets *et al.* 2014) obtained using other GACs. This is likely to be primarily associated with the higher specific surface area (and hence with the higher availability of adsorption sites) of the GAC used for our experiments ($774 \text{ m}^2 \text{ g}^{-1}$) compared to that used in the earlier studies (560 and $500 \text{ m}^2 \text{ g}^{-1}$, respectively). However, the observed increase in the adsorption capacity is not strictly proportional to the increase in the specific surface area, suggesting that other factors, such as the PZC, may also play a relevant role in the adsorption process. The PZC indicates the pH at which a surface has a net neutral charge. Although metaldehyde is a neutral compound, its oxygen atoms bear a negative partial charge that may strongly influence the adsorption behaviour of the pesticide depending on the surface charge of the adsorbent. Surfaces with low PZC normally exhibit a negative charge at environmental pH in contrast to those having high PZC, which are positively charged over a wider pH range. Based on the above considerations, metaldehyde is expected to be preferentially adsorbed on GAC with higher PZC. In line with this, it was found that the GAC used in the present work has a PZC value of 8.7 (see Figure 6), a value sufficiently high to assure a high metaldehyde adsorption efficiency (Busquets *et al.* 2014).

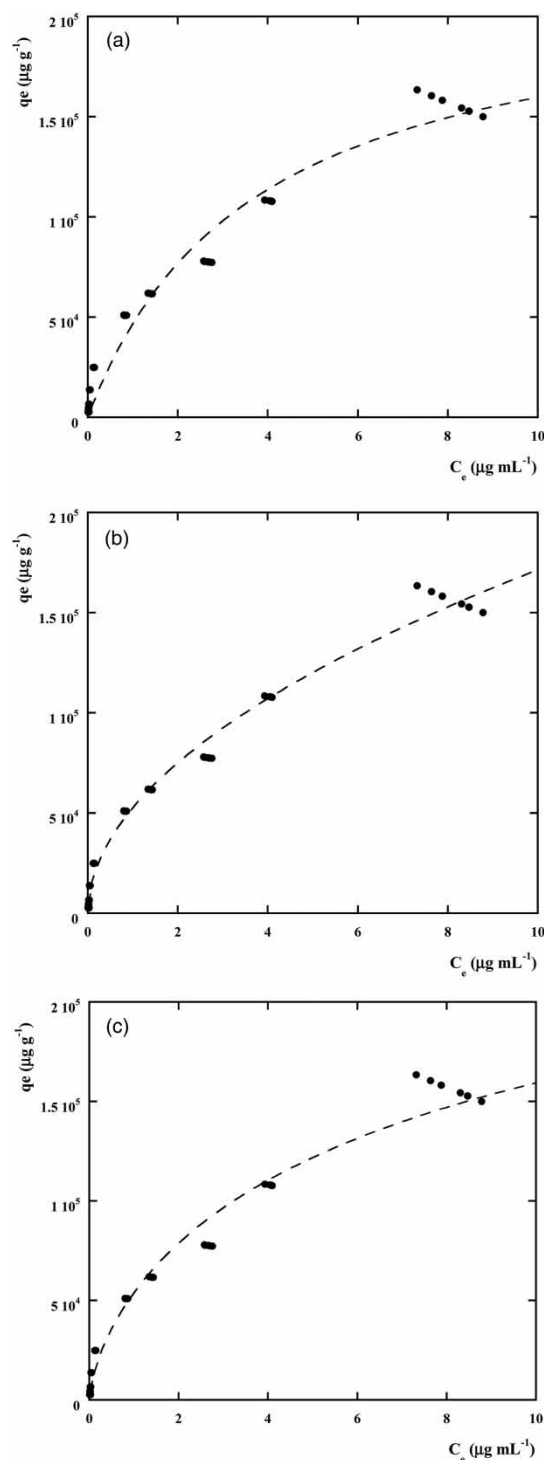
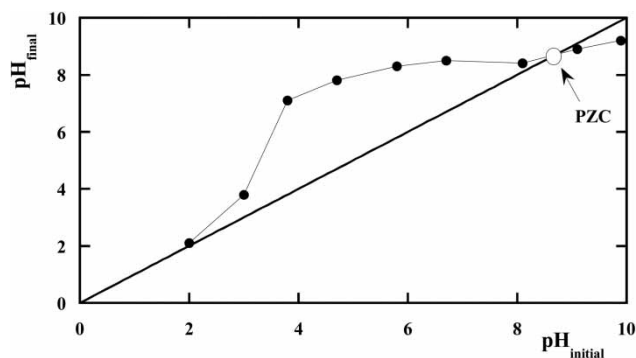


Figure 5 | Sorption isotherms of metaldehyde determined according to the following models: (a) Langmuir model; (b) Freundlich model; (c) Langmuir-Freundlich model.

It is also interesting to note that the sorbent used here has a higher affinity for metaldehyde, as the initial slope of

Table 3 | Thermodynamic parameters as determined by the fitting procedure

Model	K_L (L mg ⁻¹)	K_F (mg ^{1-N} g ⁻¹ L ^N)	K_{LF} (L ^N mg ^{-N})	N	q_{max} (mg g ⁻¹)	R^2
Langmuir isotherm	270 ± 30				220 ± 10	0.962
Freundlich isotherm		1,800 ± 30		0.51 ± 0.10		0.989
Langmuir–Freundlich isotherm			$(1.7 ± 0.4) × 10^{-3}$	0.69 ± 0.06	(320 ± 70)	0.980

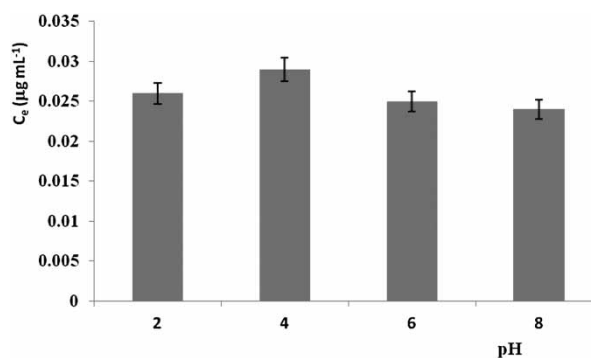
**Figure 6** | Determination of PZC for GAC.

its isotherm is greater than that of the other GACs (Tao & Fletcher 2013; Busquets *et al.* 2014).

As mentioned in the introduction section, one of the main issues is the inefficiency of water treatment plants for reducing the metaldehyde concentration below the European limit of 0.1 µg L⁻¹. Based on our results and considering that treatment plant effluents may contain up to 1.03 µg L⁻¹ of metaldehyde (Busquets *et al.* 2014), it can be estimated that each gram of GAC would be sufficient to treat about 17,000 L of waste water in batch reactors so as to bring the concentration below the legal limit.

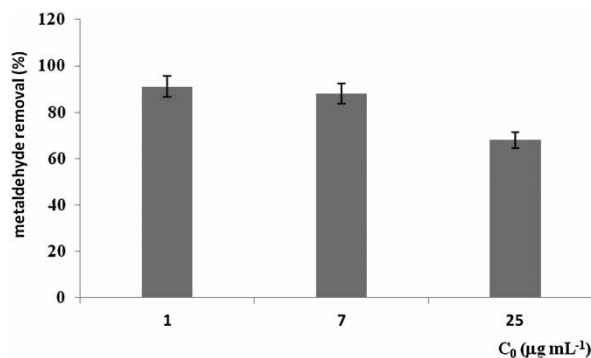
Effect of pH, ionic strength and initial concentration

In order to investigate the effect of pH on the sorption of metaldehyde onto GAC, some experiments were carried out by varying pH in the range of 2–8. The results reported in Figure 7 indicate that pH has no significant influence on the sorption of metaldehyde onto GAC. Notably, a few experiments carried out in water at pH = 2 revealed that metaldehyde is fully hydrolyzed to acetaldehyde, in agreement with previous findings (Booze & Oehme 1986). This indicates that the presence of GAC has an inhibitory effect on the acidic hydrolysis of metaldehyde.

**Figure 7** | Effect of pH on metaldehyde equilibrium concentration.

The results of the sorption experiments obtained by varying the initial solute concentration (C_0) are showed in Figure 8. As expected, the removal efficiency decreased with increasing C_0 . In particular, the removal efficiency was very high (91%) at the lowest dose tested (1 ppm), whereas it decreased significantly, up to 68%, when C_0 was 25 ppm.

To understand the effect of ionic strength, the sorption performance of GAC in the presence of KCl was examined. The sorption isotherms (not shown), within the experimental errors, were quite similar in the range of concentration explored (0–1 M). Therefore, ionic strength has no influence on the sorption process.

**Figure 8** | Effect of the initial liquid phase concentration on metaldehyde sorption.

Likewise, the temperature also has no detectable influence on the sorption isotherms ($\Delta H^\circ \approx 0$), hence suggesting that sorption is physisorption (Colella et al. 2015).

CONCLUSIONS

In the present work, the sorption of metaldehyde onto GAC was investigated. It was found that GAC has an energetically heterogeneous surface. The rate of the sorption is diffusion-controlled. Among different models tested, the hybrid Freundlich isotherm was found to describe more adequately the sorption data at equilibrium, whereas the Vermeulen equation was found to be appropriate for modelling kinetic data, indicating that the process is rate limited by intraparticle diffusion. Metaldehyde uptake is likely promoted by electrostatic and/or H-bond interactions between the electro-negative oxygen atoms of the molecule and the positively charged surface of the adsorbent. Ionic strength, pH and temperature have no significant influence on metaldehyde uptake.

ACKNOWLEDGEMENTS

The authors are grateful to Dr Judith Zhou and Dr Catherine Unsworth of the Department of Civil, Environmental and Geomatic Engineering at University College London, for their precious technical support. Part of this study was supported by the DST-UKIERI Thematic Partnerships (DST-2013-14/080).

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