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Prediction of powdered activated carbon doses for 2-MIB removal in drinking water treatment using a simplified HSDM approach

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HIGHLIGHTS

- We propose a simplified approach to quickly determine PAC dose for 2-MIB removal.
- After simplification, HSDM parameters were obtained by only one kinetic experiment.
- The Freundlich 1/n value for the approach was assumed to be in a range of 0.2–0.6.
- The simplified approach well predicts the data for 5 PACs in 3 source waters.
- The approach was further validated with the data collected from one full scale WTP.

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ABSTRACT

The addition of powdered activated carbon (PAC) is an effective measure to cope with seasonal taste and odor (T&O) problems caused by 2-methylisoborneol (2-MIB) and *trans*-1, 10-dimethyl-*trans*-9-decalol (geosmin) in drinking water. Some T&O problems are episodic in nature, and generally require rapid responses. This paper proposed a simplified approach for the application of the homogenous surface diffusion model (HSDM) to predict the appropriate PAC doses for the removal of 2-MIB. Equilibrium and kinetic experiments were performed for 2-MIB adsorption onto five PACs in three source waters. The simplified HSDM approach was compared with the experimental data, by assigning the Freundlich 1/n value in the range of 0.1–1.0 and obtaining the Freundlich equilibrium parameter K value through a 6-hr adsorption kinetic test. The model describes the kinetic adsorption data very well for all of the tested PACs in different source waters. The results were validated using the data obtained from one full scale water treatment plant, and the differences between the predicted and observed results were within 10% range. This simplified HSDM approach may be applied for the rapid determination of PAC doses for water treatment plants when faced with 2-MIB episodes in source waters.

1. Introduction

Lake and reservoir source waters often experience periodic odor episodes (Suffet et al., 1996; Watson et al., 2008; Watson, 2003; 2004). Although this is typically an aesthetic issue, water companies often have to invest considerable amounts of money in response to customer complaints caused by the resulting taste and odor (T&O) problems (Suffet et al., 1996). 2-Methylisoborneol (2-MIB) and *trans*-1, 10-dimethyl-*trans*-9-decalol (geosmin), two secondary metabolites of some cyanobacteria and actinomycetes (Wilkins, 1996; Zaitlina and Watson, 2006; Wu and Duirk, 2013), are the most frequently encountered odorants in drinking water. These two compounds can impart an earthy/musty odor, may be perceived even at levels as low as 10 ng/L (Watson et al., 2000), and are not readily removed by conventional water treatment processes (Lin et al., 2002; Ho et al., 2007, 2012; Zamyadi et al., 2015). However addition of powdered activated carbon (PAC) has been demonstrated to be an effective approach to dealing with the

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periodic odor problems in drinking water caused by 2-MIB/geosmin (Lalezary-Craig et al., 1988; Cook et al., 2001; Matsui et al., 2009).

Adsorption of 2-MIB/geosmin onto PACs in natural water strongly depends on the properties of activated carbon used (Lalezary-Craig et al., 1988; Pendleton et al., 1997; Newcombe et al., 2002; Yu et al., 2007) and the compositions of natural organic material (NOM) in the water (Lalezary-Craig et al., 1988; Chen et al., 1997; Newcombe et al., 1997). The properties of PAC, including surface functional groups and pore structure have been considered as the factors influencing the adsorption capacity of trace organic compounds onto PACs (Lahaye, 1998; Radovic et al., 1997). More recently, micropore volume has been proposed to be an effective indicator to estimate the adsorption for 2-MIB/geosmin onto PAC (Newcombe et al., 2002; Nowack et al., 2004; Yu et al., 2007). NOM is known to suppress the adsorption capacity of targeted organic compounds onto PAC during adsorption processes (Newcombe et al., 1997). For water treatment scenarios, the concentrations of contaminants of interest such as 2-MIB/geosmin and atrazine are at ng/L to μg/L level, and are significantly lower than that of NOM, usually at mg/L level. Under this condition, the removal ratios of 2-MIB/geosmin by PAC in NOM-laden natural water under equilibrium condition was found to be independent of initial 2-MIB/geosmin concentration and only dependent on carbon dose (Gillogly et al., 1999; Graham et al., 2000). This relationship could be simulated with bi-solute ideal adsorbed solution theory (IAST) (Knappe et al., 1998; Graham et al., 2000), in which NOM and the targeted compound are considered as two solutes.

In water treatment plants, the typical contact times of PAC are in the order of 1 h, which is much less than the equilibrium time for PAC adsorption. Therefore, adsorption kinetics should be considered for PAC application. Although the observation of 2-MIB/geosmin removal ratio being independent of initial concentration has been extended to non-equilibrium conditions for PAC adsorption in natural water (Matsui et al., 2001, 2002, 2003; Zoschke et al., 2011), to determine an appropriate PAC dose, multiple experiments are needed for different PAC doses for a specific natural water. Homogeneous surface diffusion model (HSDM), developed based on the diffusion of adsorbed adsorbate molecules on the surface of internal pores within adsorbent particles (Crittenden and Weber, 1978), has been successfully employed to simulate the adsorption kinetics of a range of compounds, including 2-MIB/geosmin onto activated carbon (Sontheimer et al., 1988; Najm et al., 1991; Huang et al., 1996; Gillogly et al., 1998). The model makes it possible to predict the carbon doses required for drinking water treatment and has been tested in several studies (Najm et al., 1991; Huang et al., 1996; Gillogly et al., 1998, 1999; Graham et al., 2000). Gillogly et al. (1998) studied the adsorption 2-MIB onto five PACs in Lake Michigan water, and demonstrated that the PAC doses required for any contact time of interest may be quickly determined with the HSDM. Similarly, Cook et al. (2001) successfully employed the HSDM to predict the PAC doses required to reduce 2-MIB and geosmin concentrations to below 10 ng/L in three of the four waters in Adelaide, South Australia. The only poorly predicted case was attributed to higher turbidity in the studied water, which made PAC incorporate into a larger, denser floc, leading to a reduced effective contact time of the adsorbent (Cook et al., 2001).

When applying the HSDM, the surface diffusion coefficient (D_s) and the two Freundlich isotherm constants (K and 1/n) need to be known in advance. Adsorption of 2-MIB and geosmin onto activated carbon is known to be influenced by the type of adsorbent and water matrix (Knappe et al., 1998; Summers et al., 2013). Therefore, for each source water/PAC combination, experiments are needed to obtain D_s, K, and 1/n. At least one adsorption kinetic experiment is required for the determination of D_s, and three equilibrium experiments for K and 1/n as demonstrated in previous studies (Gillogly et al., 1998; Cook et al., 2001). These adsorption tests may generally take several days to reach equilibrium, due primarily to the slow diffusion process of the adsorbates within the intraparticle pores (Gillogly et al., 1998). It is therefore desirable if the number of adsorption tests can be reduced and time required for the experiments can be shortened for a faster response. Huang (Huang et al., 1996) suggested that the three HSDM parameters, D_s, K and 1/n, could be extracted from fitting the data of three kinetic tests, which may significantly reduce the effort required for the equilibrium experiments. However, the model was only tested with one PAC and one source water. In addition, three parameters were adjustable in the model fitting, making the calculation more complicated.

Table 1 summarizes the values of 1/n for the adsorption of 2-MIB onto the PACs reported in the literature. As indicated, the values normally fall into the range of 0.1–1.0 and between 0.2 and 0.5 for almost all the cases in natural water. If assigning a 1/n value within the above range, the Freundlich K may be estimated from the adsorption capacity data obtained from the plateau values at the end of the kinetic experiments. Then D_s value is the only adjustable parameter in the model, and can be optimized through minimizing the differences between the data obtained in the kinetic test and those predicted by the model. In this study, the feasibility of this simplified approach for the use of HSDM for 2-MIB removal was tested with different PAC/source water combinations, and further verified using the field data collected from a full scale water treatment plant during a 2-MIB episode. The results of this study could provide a simple approach to quickly determine the appropriate carbon dose for controlling seasonable odor problems in waterworks.

2. Material and methods

2.1. Materials

2-MIB. 2-MIB was purchased from Sigma-Aldrich Co., USA, at a concentration of 10 mg/mL in methanol. Stock solutions of 1 mg/L were prepared by diluting the methanol solution with ultra-pure water (resistivity $\geq$18 m cm^{-1}).

Natural waters. Three natural waters collected from Taiwan (Fengshen Reservoir and Chengching Lake) and mainland China (Miyun Reservoir) were used in batch adsorption experiments. The pH was 7.5, 8.1 and 7.9, and the dissolved organic carbon was 2.8, 1.3 and 2.1 mg/L, respectively.

Powdered activated carbons. Five commercial powdered activated carbon (PACs) products were used in the adsorption experiments, including three coal-based ones: (WPH_PAC, Calgon Carbon Corp., USA; SX_PAC, Shanxi Xinhua Carbon Corp., China; NX_PAC, Ningxia Taixi Carbon Corp., China), a wood based one (TAC_PAC, Taiwan Active Carbon Industry Co.), and one used in Fengshen Water Treatment Plant (FS_PAC). Prior to batch experiments, the PAC samples were washed using ultra-pure water, dried overnight at 110 °C to remove excess water, and then cooled and stored in a desiccator. Physical characterization of the PACs was conducted using a surface analyzer (ASAP, 2010; Micromeritics, USA), and the data are shown in Table 2.

2.2. Equilibrium adsorption test

The bottle-point technique was used for the adsorption isotherm tests. Water samples containing 2-MIB were acquired by spiking the stock solution to the natural waters. PAC slurry was prepared by mixing 10 g oven-dried PAC in 1 L ultra-pure water before adding into the water samples. The carbon doses varied