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Research article

Adsorption of organic stormwater pollutants onto activated carbon from sewage sludge

Karin Björklund [a, b, *], Loretta Y. Li [a]

[a] Department of Civil Engineering, University of British Columbia, 6250 Applied Science Lane, Vancouver, B.C, V6T 1Z4, Canada
[b] Department of Civil and Environmental Engineering, Chalmers University of Technology, Sven Hultins Gata 8, 41296 Göteborg, Sweden

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ABSTRACT

Adsorption filters have the potential to retain suspended pollutants physically, as well as attracting and chemically attaching dissolved compounds onto the adsorbent. This study investigated the adsorption of eight hydrophobic organic compounds (HOCs) frequently detected in stormwater – including four polycyclic aromatic hydrocarbons (PAHs), two phthalates and two alkylphenols – onto activated carbon produced from domestic sewage sludge. Adsorption was studied using batch tests. Kinetic studies indicated that bulk adsorption of HOCs occurred within 10 min. Sludge-based activated carbon (SBAC) was as efficient as tested commercial carbons for adsorbing HOCs; adsorption capacities ranged from 70 to 2800 μg/g ($C_{initial}$ = 10–300 μg/L; 15 mg SBAC in 150 mL solution; 24 h contact time) for each HOC. In the batch tests, the adsorption capacity was generally negatively correlated to the compounds' hydrophobicity (log K_{ow}) and positively associated with decreasing molecule size, suggesting that molecular sieving limited adsorption. However, in repeated adsorption tests, where competition between HOCs was more likely to occur, adsorbed pollutant loads exhibited strong positive correlation with log K_{ow}. Sewage sludge as a carbon source for activated carbon has great potential as a sustainable alternative for sludge waste management practices and production of a high-capacity adsorption material.

1. Introduction

Municipal and industrial wastewater treatment plants produce large volumes of sludges, for which the processing and disposal are becoming complex problems. In Canada, a country with 24 million of its 35 million people connected to municipal sanitary sewers, an estimated 860,000 tonnes of dry biosolids are produced annually (LeBlanc et al., 2008). Corresponding numbers for the European Union, with 360 million inhabitants, are approximately 10 million tonnes of produced sewage sludges, a number deemed to be underestimated (Milieu Ltd, 2010). Major disposal routes of sewage sludge include incineration, landfilling and application to land (Fytili and Zabaniotou, 2008; LeBlanc et al., 2008). Incineration faces high capital costs and public concern over air emission risks, while rising land prices and strategies to ban dumping of organic materials have constrained landfilling. Recycling or further use of the sludge is the preferred and sustainable management option. Sewage sludge contains valuable organic matter and nutrients and could be used as a renewable source of fertilizers. Wastewater streams are, however, contaminated with chemicals and by-products from industry – including metals, polychlorinated biphenyls (PCBs); polycyclic aromatic hydrocarbons (PAHs); dioxins and furans – as well as compounds found in common household products – e.g. metals, flame retardants including polybrominated diphenyl ethers (PBDEs); plasticizers such as phthalates; surfactants including alkylphenols; personal care products and pharmaceuticals – which risk being enriched in sewage sludge (Bright and Healey, 2003; Harrison et al., 2006). The occurrence of contaminants in sludge has led to negative public perception about biosolids and is currently a major deterrent to agricultural application of sewage sludges in many countries (LeBlanc et al., 2008).

Since sewage sludge is a carbon-rich, renewable and vast resource which can be obtained at low cost, an alternative use of sludge is for the production of activated carbon (AC) adsorbents, used to remove a wide range of contaminants from air and water. Sludge has been shown to produce high-quality carbons for adsorption of impurities in water, including metals, phenols and dyes (Smith et al., 2009; Xu et al., 2015). Producing AC from sludge

* Corresponding author. Department of Civil and Environmental Engineering, Chalmers University of Technology, Sven Hultins Gata 8, 41296 Göteborg, Sweden
E-mail address: karin.bjorklund@chalmers.se (K. Björklund).

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has the potential to be a cost-effective alternative with respect to both waste management and production of low-cost adsorbents. In fact, production costs of sludge-based carbon, which depend on factors such as sludge availability and required processing, including energy costs for pyrolysis and drying, have the potential to be on par with, or even lower than, production costs of commercial ACs (Xu et al., 2015).

The pure AC carbon structure is non-polar and accordingly, ACs have shown high adsorption capacity for many types of non-polar or slightly polar organic compounds, including phenolic compounds (Liu et al., 2010), pesticides, PCBs (Sotelo et al., 2002), phthalates (Venkata Mohan et al., 2007), and PAHs (Valderrama et al., 2008). The hydrophilicity of ACs can be increased by the presence of polar surface functional groups, and the surface chemistry can be modified to enhance selectivity of specific chemicals. Inorganics, including for example Cu, Pb and Zn (Rivera-Utrilla et al., 2011), nitrate, nitrite and ammonia (Afkhami et al., 2007; Huang et al., 2008), as well as sulfides, chlorides and cyanides (Bansal and Goyal, 2005), have been shown to adsorb onto modified ACs.

Activated carbon is regularly used in drinking water and wastewater treatment, but currently has limited application for other types of contaminated water, including stormwater, which is often contaminated with metals, organic pollutants and nutrients. Recent research shows that many persistent organic pollutants, including PAHs, PCBs, PBDEs, phthalates and alkylphenols, are ubiquitous in stormwater (Björklund et al., 2009, 2011; Bressy et al., 2012; Zgheib et al., 2011). Stormwater contaminated with organic substances may pose a threat to receiving waters, as many organic compounds have been shown to persist in the environment, bioaccumulate in organisms, and exhibit adverse effects on human and animal health. To achieve established water quality objectives, stormwater treatment is considered critical.

Contrary to what has been assumed previously, recent studies show that many theoretically hydrophobic organic compounds (HOCs) with high water-octanol partition coefficient (K_{ow}), such as PAHs and phthalates, are found in considerable amounts in the dissolved (<0.7 μm) and colloidal phases of stormwater (Kalmykova et al., 2013; Zgheib et al., 2011). Consequently, stormwater treatment methods, such as filtration and sedimentation, are assumed to be inefficient for HOCs in non-particulate phases. Adsorption filters have the potential to physically retain suspended pollutants, as well as attracting and chemically attaching dissolved pollutants onto adsorbent surfaces. Adsorption can be adopted in stormwater treatment methods such as storm drain filters and bioretention. Provided that effective media are used, adsorption can be an economical and easily-operated treatment method. Hence, AC produced from sewage sludge has the potential to become a sustainable solution for improving stormwater and receiving water quality.

The objectives of this study were to i) investigate the adsorption of organic stormwater contaminants onto sludge-based carbon in the absence and presence of competing adsorbates; ii) compare the adsorption capacity of sludge-based activated carbon (SBAC) with a surface-modified SBAC and two commercially available ACs. This is the first evaluation of the adsorption efficiency of SBAC for HOCs; adsorption is investigated in batch tests using ultrapure water spiked with organic compounds. Organic contaminants receive particular attention, as there are currently few data on the use of adsorption to remove them from stormwater (Li and Helmreich, 2014; Vesting et al., 2015). Selected organic compounds – four PAHs (fluorene, anthracene, pyrene and benzo[e]pyrene), two phthalates (dibutyl and di(2-ethylhexyl) phthalate), as well as two alkylphenols (4-*t*-octyl- and 4-nonylphenol) – were chosen according to reported stormwater detection frequency (Björklund et al., 2011; Bressy et al., 2012; Zgheib et al. 2011, 2012) and contrasting characteristics such as hydrophobicity and functional groups (Table S1, Supplementary Material).

2. Material and methods

2.1. Production of sludge-based activated carbon

The preparation of SBAC has been described in detail elsewhere (Gong, 2013), and is only summarized here. Sludge was collected from the aerobic zone of the wastewater treatment pilot plant at the University of British Columbia, Vancouver, which receives domestic wastewater from the campus. Because sludge quality (e.g. water and organic content, metal concentrations) vary over time, the sludge was collected at several occasions to avoid having a final carbon material that reflects only a limited quality of sludge. The sludge was centrifuged, dried (105 °C) and ground to a fine powder. The powdered sludge was soaked in 5 M $ZnCl_2$ for 24 h, dried for 24 h, then pyrolysed at T_{final} = 500 °C in an electric furnace. After conversion, the SBAC was ground to a powder (50–100 mesh), washed with 5 M HCl followed by distilled water, dried and stored in amber glass bottles.

Physical and chemical characterization of the produced carbon materials included carbon content, BET surface area, pH, cation exchange capacity (CEC), X-Ray Diffraction, and surface functional groups by Fourier transform infrared spectroscopy (FTIR), all explained in detail by Gong (2013).

2.2. Other sorbents

For comparison, SBAC modified with an oxidizing agent was tested. A pre-study (Gong, 2013) showed that impregnating SBAC with 10 M HNO_3 (1 g–10 mL) at 90 °C for 4 h produced a modified SBAC (modSBAC) with excellent adsorption capacity for metals, and hence this was tested in this study. Carbonized sludge (CS) was prepared by pyrolysing the sludge using the same method as for the SBAC, but without $ZnCl_2$ impregnation.

Two commercially available ACs from Sigma Aldrich (untreated powder, 100–400 mesh, henceforth referred to as Sigma) and Calgon Carbon (powdered, WPH type for water treatment, 100–325 mesh, henceforth referred to as WPH) were tested in parallel with the carbons produced from sewage sludge.

2.3. Batch adsorption tests

2.3.1. Adsorption isotherms of organic compounds

Two solutions were prepared for isotherm tests of HOCs: (1) individual organic compounds spiked at five concentrations (10, 50, 100, 200 and 300 μg/L), and (2) a mixture of all eight compounds, each spiked at the same concentrations range as (1) above. Higher concentrations could not be tested due to the limited water solubility of HOCs. In amber glass bottles, 150 mL ultrapure water was spiked with HOCs, the solution was thoroughly mixed, and 0.015 g of each the four ACs (SBAC, modSBAC, WPH, Sigma) were added. Samples were mixed using an end-over-end rotator at room temperature (20 ± 2 °C) for 24 h, then centrifuged at 2000 rpm for 10 min: the liquid phase was analyzed for concentrations of HOCs. Samples with individual compounds spiked were prepared in triplicate; all other samples were prepared in duplicate. Blanks of all carbons (SBAC, modSBAC, CS, WPH and Sigma) in water were analyzed to determine leaching of organic compounds and metals.

For each batch of tests, a matrix blank and a matrix spike were prepared following the same procedure as for the adsorption tests to determine contamination and loss of analytes, respectively. Matrix spikes showed acceptable recoveries (>70%) and good