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Review

Performance of granular activated carbon to remove micropollutants from municipal wastewater—A meta-analysis of pilot- and large-scale studies

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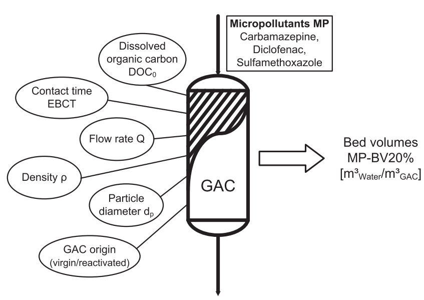
HIGHLIGHTS

- Adsorbers operating time differs by 2500% until diclofenac-BV20% was reached.
- Elimination of diclofenac even when equilibrium of DOC was reached.
- No strong statistical significance of EBCT and DOC_0 on MP-BV20% due to lack of data and high heterogeneity of studies.
- Adsorbers should be operated ≫20,000 BV for exact calculation of breakthrough curves.
- We generally recommend using reactivated GAC and to carry out pilot tests.

GRAPHICAL ABSTRACT



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ABSTRACT

For reducing organic micropollutants (MP) in municipal wastewater effluents, granular activated carbon (GAC) has been tested in various studies. We did systematic literature research and found 44 studies dealing with the adsorption of MPs (carbamazepine, diclofenac, sulfamethoxazole) from municipal wastewater on GAC in pilot- and large-scale plants. Within our meta-analysis we plot the bed volumes (BV $[m^3_{water}/m^3_{GAC}]$) until the breakthrough criterion of MP-BV20% was reached, dependent on potential relevant parameters (empty bed contact time EBCT, influent DOC DOC_0 and manufacturing method). Moreover, we performed statistical tests (ANOVAs) to check the results for significance. Single adsorbers operating time differs i.e. by 2500% until breakthrough of diclofenac-BV20% was reached (800–20,000 BV). There was still elimination of the "very well/well" adsorbable MPs such as carbamazepine and diclofenac even when the equilibrium of DOC had already been reached. No strong statistical significance of EBCT and DOC_0 on MP-BV20% could be found due to lack of data and the high heterogeneity of the studies using GAC of different qualities. In further studies, adsorbers should be operated ≫20,000 BV for exact calculation of breakthrough curves, and the following parameters should be recorded: selected MPs; DOC_0; UVA_{254}; EBCT; product name, manufacturing method and raw material of GAC; suspended

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solids (TSS); backwash interval; backwash program and pressure drop within adsorber. Based on our investigations we generally recommend using reactivated GAC to reduce the environmental impact and to carry out tests on pilot scale to collect reliable data for process design.

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1. Introduction

In addition to having total natural organic matter (NOM), suspended solids (TSS) and nutrients, municipal wastewater contains a wide range of synthetic organic chemicals. Their origin and properties can differ significantly and can comprise pharmaceutic compounds, surfactants, inhibitors, complexing agents, sugar substitutes, pesticides of urban or domestic use, etc. (Petrie et al., 2015). A variety of these organic micropollutants (MP) and their metabolites in the wastewater are insufficiently removed since they are poorly biodegradable in conventional wastewater treatment processes (WHO, 2011). Apart from direct discharge at production sites, the secondary or tertiary effluents from municipal wastewater treatment plants (WWTP) are, thus, the most important pathway for these substances to enter the aquatic environment (Petrie et al., 2015).

Furthermore, the continuous use of MPs places a relatively uniform load on rivers and their reservoirs in a concentration range of $ng \cdot L^{-1}$ to $\mu g \cdot L^{-1}$. (Merkel, 2004; Bergmann et al., 2008; Kümmerer, 2008; Abegglen and Siegrist, 2012). In turn, this can chronically impair water biocoenosis (Suter and Holm, 2004; Galus et al., 2013). In Switzerland, one nationwide study shows that water quality requirements for many compounds cannot be met downstream of wastewater treatment plants. This, in particular, applies to water bodies that have a high proportion of wastewater (Gälli et al., 2009). Requirements that define the water quality are constantly being (re)formulated (Carvalho et al., 2015) as we gain new insight into the effects that these various substances have.

Different technologies are available for MP removal. Oxidative processes such as ozonation of WWTP effluent as well as adsorptive treatment with powdered activated carbon (PAC) have proven to be effective and feasible from a technical and economical perspective (Metzger, 2010; Abegglen and Siegrist, 2012; Böhler et al., 2012b). Several large-scale plants for ozonation and PAC treatment have already been built in Switzerland and Germany. In addition, the use of granular activated carbon (GAC) is considered to be a viable method for eliminating MPs. GAC has been successfully used for decades in the field of drinking water production (Sontheimer et al., 1988; Oxenford and Lykins, 1991), ground water remediation (Culver and Shenk, 1998) and industrial wastewater treatment (Kienle and Bäder, 1980; Bansal and Goyal, 2005).

Owing to its very large inner surface area (typically of 800 to1,200 m^2 g^{-1}) (Kienle and Bäder, 1980; Sontheimer et al., 1988), activated carbon has a high capacity for adsorbing dissolved organic substances from surrounding water. GAC performance depends on the raw material used for its production (bituminous coal, lignite, coconut shell, wood, peat, synthetic polymers) (Kienle and Bäder, 1980), on the way this material was initially thermally carbonized during production (for removal of volatiles) and on the subsequent thermal activation (i. e. specific burn-off of carbon to increase the inner surface area) (DIN EN 12915-2, 2008; DIN EN 12915-1, 2009). Some special forms of GAC are re-agglomerated from PAC. All of these factors influence the internal grain structure, as well as the size and the physico-chemical properties of the inner surface, which consists of inhomogeneous microcrystalline graphite layers (Kienle and Bäder, 1980; Sontheimer et al., 1988). GAC grains of have diameters in the range of a few millimetres (typically 0.5–4.0 mm) and are used in adsorbers (packed bed filters) (Tchobanoglous et al., 2003; DIN EN 12915-2, 2008; DIN EN 12915-1, 2009; ANSI/AWWA B604-12, 2012).

Once the GAC is loaded, it must be thermally reactivated or replaced by virgin GAC. For reactivation the exhausted GAC is hydraulically transferred from the adsorber into silo trucks and transported to a reactivation plant (Benstöm et al., 2014b). Since the grains of exhausted GAC are still filled with water, it is commonly fed through a rotary kiln or multiple hearth furnace that provides different sections for grain drying at low temperature and desorption of volatile organic compounds and pyrolysis at approx. 800 °C (Henning and Wanzl, 2008). The inevitable loss of GAC during the whole reactivation process caused by burn-off, abrasion and sieving is typically 5–15% (Grombach et al., 2000). The amount of loss depends on the type of GAC, its preceding use in the specific adsorption process (i. e. organic and inorganic load, operation time) and the operation of the reactivation furnace (Kienle and Bäder, 1980; Sontheimer et al., 1988). To compensate for this loss, virgin GAC has to be added – so-called “make-up”. The main share of these losses (abrasion and pass through of sieving) for GAC makes its way as powder activated carbon back into the market (Alt et al.,