

Direct dosage of reactivated carbon from waterworks into the activated sludge tank for removal of organic micropollutants

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ABSTRACT

The thermal reactivation of granular activated carbon is a substantial ecological and economic benefit in the process of drinking water treatment. A significant amount of abraded carbon, which is similar to powdered activated carbon (PAC), is produced that can be brought to application at wastewater treatment plant level for the removal of micropollutants in a powdered activated carbon-activated sludge (PAC-AS) system. This excess carbon derived as a by-product from the reactivation process in a waterworks was applied directly into the activated sludge tank and has been elevated in this study by monitoring the removal efficiencies for benzotriazole, carbamazepine, diclofenac, metoprolol and sulfamethoxazole in the effluent of a semi-technical wastewater treatment plant of 39 m³. A simulation-derived sampling strategy was applied to optimize the recovery rates of the micropollutants. Flow-proportional, 72-hour composite sampling was considered best. The elimination rates obtained for a 10 g PAC·m⁻³ dosage were 73% for benzotriazole, 59% for carbamazepine, 60% for diclofenac, 67% for metoprolol and 48% for sulfamethoxazole. The obtained results underline the importance of appropriate sampling strategies, which can be derived from hydraulic modeling.

Key words | flow-proportional sampling, hydraulic modeling, micropollutants, PAC-AS, reactivated carbon, time-proportional sampling

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INTRODUCTION

Numerous micropollutants, such as pharmaceutical products, endocrine disruptors or hormones, are present in our water bodies with wastewaters having the highest variety at trace concentrations, ranging from µg·m⁻³ to mg·m⁻³. Since in practice conventional wastewater treatment plants (WWTPs) are not designed to eliminate micropollutants as a foremost priority, advanced treatment steps, such as ozonization, adsorption on powdered activated carbon (PAC), or filtration through granular activated carbon (GAC), have proven to be very efficient approaches (Mousel *et al.* 2017). Among these, PAC combined with sand filtration is probably the most widespread method, yielding efficiencies of up to 80–95%, with a relative low energy demand of 0.03 kWh_{el}·m⁻³ for a dosage of 10 g PAC·m⁻³ respectively (Lowenberg *et al.* 2014; Mousel *et al.* 2017).

Nowadays, a variety of biogenic materials and waste products with sufficiently high carbon content and low

percentage of inorganics are suitable as raw materials for activated carbon production. Those comprise for instance palm shells, coconut coir pith, olive waste, rice husk, coffee extract residues or coconut shells (Arena *et al.* 2016). All these raw materials are beneficial in terms of their economic viability or carbon footprint.

At waterworks activated carbon is commonly used for the removal of dissolved organics from drinking water. Due to economic reasons, exhausted carbons are mostly thermally reactivated and reused. During this process, 10–15% of carbon is ‘lost’ due to abrasion, which sums up to roughly 400 tons per year at the Düsseldorf-Holthausen waterworks. This amount could potentially act as a substitute for fresh activated carbon during advanced wastewater treatment. At the plant site there are few options available as to how PAC is applied. The powdered activated carbon-activated sludge (PAC-AS) method implies the direct

dosing of PAC into the activated sludge treatment unit of the plant. The advantageous effect of direct PAC application was recently demonstrated (Aziz *et al.* 2011; Hu *et al.* 2015; Karelid *et al.* 2017), but to the authors' knowledge there has been no focus on pharmaceutical removal in PAC-AS systems at full-scale level. Aziz *et al.* (2011) were able to demonstrate the superior performance regarding the removal efficiencies for chemical oxygen demand, color, ammonia nitrogen, and total dissolved salts, which was later confirmed by Hu *et al.* (2015). Moreover, PAC-AS systems will also benefit the performance of the involved microorganisms as the direct PAC dosing will protect them from toxic substances in the wastewater matrix such as heavy metals and from substances having an inhibitory effect (Kuai *et al.* 1998; Mochidzuki & Takeuchi 1999; Hu *et al.* 2015). The dosing of the PAC can be performed according to the individual needs, yielding rapid adsorption kinetics. PAC-AS systems also require a significantly lower amount of carbon in comparison to GAC filters, and these filters also lose their sorption capacity over time due to dissolved organic carbon loading, resulting in a filter breakthrough with increasing micropollutant effluent concentrations.

The direct dosing of the excess carbon from waterworks into the activated sludge tank of a WWTP would offer a relatively simple to implement alternative to construction measures for a fourth treatment stage. This is especially advantageous for municipalities that operate both waterworks and WWTPs as no external purchase of activated carbon is necessary anymore.

Nonetheless, new technical procedures always require an appropriate monitoring campaign in order to prove their applicability. Attention should always be paid to the sampling strategy that is at the front in monitoring campaigns and which is the major source of inaccuracy, especially as modern chemical analysis methods are able to detect water constituents at very low levels (Ort *et al.* 2010). In order to get a representative and reliable micropollutant concentration or load, the sampling strategy has to be optimized according to the substance's temporal load pattern, the sampling interval and the length of the composite sampling time (Ort & Gujer 2006). In the majority of publications it is rarely accurately described how sampling was performed (Ort & Gujer 2006). Under certain circumstances this brings up a challenge when comparing the different outcomes of published studies as monitored results are subject to huge fluctuations, which can be seen in the review by Zhang *et al.* (2008) for the example of the removal efficiencies of carbamazepine and diclofenac. Currently, 24-hour composite sampling is often performed,

whereby the effluent sampling is shifted by the mean hydraulic retention time, which disregards mixing regime characteristics as well as flow and concentration variability (Clara *et al.* 2005; Gobel *et al.* 2007; Majewsky *et al.* 2011). In consequence, huge fluctuations, biased estimates or negative elimination rates might be the outcome as indicated in recent publications. Ort *et al.* (2010) were able to show that depending on the sampling strategy the concentrations of iopromide and carbamazepine varied between 11–15 mg·m⁻³ and 1.1–1.6 mg·m⁻³, while negative elimination rates for micropollutants were found in studies by Clara *et al.* (2004), Vieno *et al.* (2007), and Le-Minh *et al.* (2010).

The derivation of an appropriate sampling strategy demands a powerful tool at hand that is capable of assessing the micropollutants' hydraulic residence times at WWTP level. Dynamic models have already been proven to be a robust solution for various scientific questions and for many different WWTPs (Wichern *et al.* 2003). Hence, dynamic models might meet this demand. However, these models mostly imply biological models. As micropollutants can be seen as inert compounds, these models can be reduced to hydraulic models simulating their transport and mixing processes only. Thus, it will be possible to assess their real hydraulic residence times from which an appropriate sampling strategy can be derived.

The overall aim of this study was to demonstrate the advantage of using excess carbon from a waterworks for direct application in an activated sludge tank of a WWTP for the removal of pharmaceuticals. The study was supported by deriving an optimized sampling strategy for the investigated micropollutants by means of hydraulic modeling. The applicability of hydraulic modeling for sample strategy derivation and the use of reactivated PAC was shown at a semi-technical WWTP for the example of the elimination of five selected micropollutants that were benzotriazole, carbamazepine, diclofenac, metoprolol and sulfamethoxazole.

METHODS

Semi-technical wastewater treatment plant

The semi-technical WWTP (500 population equivalent) is situated within the site of the wastewater treatment plant Düsseldorf-Süd WWTP (Stadtentwässerungsbetrieb Düsseldorf, Germany). The pilot plant consists of two independent, but identical activated sludge treatment tanks with their connected secondary clarifiers. One treatment line was used for testing, while the other one was used as a reference line. The

total volume of each activated sludge tank is 39 m^3 , of which 50% was used for preliminary denitrification. Aeration during nitrification was performed with membrane disc diffusers (WILO GVA GmbH, Wülfrath, Germany) maintaining a dissolved oxygen level of approximately $2 \text{ g}\cdot\text{m}^{-3}$. The secondary clarifiers had a total volume of 6.5 m^3 each. Generally, the semi-technical plant was operated in a way that was similar to the full-scale Düsseldorf-Süd WWTP. The plant had a constant flow of $1.3 \text{ m}^3\cdot\text{h}^{-1}$, while the return ratio of the internal recirculation was set at 4. The flow rate of the recirculation was $5.2 \text{ m}^3\cdot\text{h}^{-1}$ and the one for the return sludge was $1.3 \text{ m}^3\cdot\text{h}^{-1}$ yielding a return ratio of 1. The influent wastewater was characterized by a total organic carbon (TOC) concentration of $180 \text{ g}\cdot\text{m}^{-3}$, a total bound nitrogen (TN_b) concentration of $69 \text{ g}\cdot\text{m}^{-3}$ and by a total phosphorus (TP) concentration of $7.4 \text{ g}\cdot\text{m}^{-3}$ on average. The mixed liquid suspended solids content was $2.5 \text{ kg}\cdot\text{m}^{-3}$ and the solids retention time was between 17 and 25 days. The mean effluent levels of the test line were $0.4 \text{ g}\cdot\text{m}^{-3}$ $\text{NH}_4\text{-N}$, $4.3 \text{ g}\cdot\text{m}^{-3}$ $\text{NO}_3\text{-N}$, $2.0 \text{ g}\cdot\text{m}^{-3}$ TP and $13.1 \text{ g}\cdot\text{m}^{-3}$ TOC. For the reference line the levels were $0.4 \text{ g}\cdot\text{m}^{-3}$ $\text{NH}_4\text{-N}$, $3.6 \text{ g}\cdot\text{m}^{-3}$ $\text{NO}_3\text{-N}$, $2.5 \text{ g}\cdot\text{m}^{-3}$ TP and $14.1 \text{ g}\cdot\text{m}^{-3}$ TOC. The operating temperature range during the monitoring campaign was between 289.15 K and 297.15 K.

During two measurement campaigns, reactivated carbon (PAC) was dosed directly into the activated sludge tank of the testing line in concentrations of $10 \text{ g}\cdot\text{m}^{-3}$ and $20 \text{ g}\cdot\text{m}^{-3}$. The PAC was taken from the Düsseldorf-Holthausen drinking water treatment facility (Stadtwerke Düsseldorf AG) and produced from losses in the form of combustion and abrasion products that arise during thermal reactivation of carbon granules.

Monitoring of micropollutants in the influent and effluent

Within the present study five micropollutants have been monitored: the industrial chemical benzotriazole ($\text{C}_6\text{H}_5\text{N}_3$), the antiepileptic drug carbamazepine ($\text{C}_{15}\text{H}_{12}\text{N}_2\text{O}$), the anti-inflammatory drug diclofenac ($\text{C}_{14}\text{H}_{11}\text{Cl}_2\text{NO}_2$), the beta-blocker metoprolol ($\text{C}_{15}\text{H}_{25}\text{NO}_3$) and the antibiotic drug sulfamethoxazole ($\text{C}_{10}\text{H}_{11}\text{N}_3\text{O}_5\text{S}$). The sampling strategy for these micropollutants was derived from the simulation results within this study.

Hydraulic modeling

Dynamic modeling was performed using the Matlab[®]/Simulink[™] based simulator SIMBA 6.5 (ifak – Institut für Automation und Kommunikation e.V., Magdeburg,

Germany). A hydraulic model was developed that explicitly included transport and mixing processes to simulate the residence-time distribution (RTD) of a micropollutant in the influent and effluent of the semi-technical plant. The simulation was based on a pulse input, where a certain amount of the tracer is injected into the feed of the WWTP. The tracer outlet concentration is calculated as a function of time given an effluent concentration–time curve that is referred to as the C-curve in RTD analysis. The cumulative distribution of the tracer concentration in the effluent of the WWTP is described by the F-curve. Both curves were used as the basis in the calibration process of the hydraulic model.

The simulation was used to conduct four different scenarios, whereby in the first three scenarios different pulsed peaks in the influent were tested. The first scenario dealt with a single concentration peak in the influent, the second scenario with two single peaks and the third scenario with a constant concentration over 24 hours. All three scenarios were simulated at a dry weather flow of $Q_d = 1.3 \text{ m}^3\cdot\text{h}^{-1}$, for the case of a rain event with $Q_r = 2.6 \text{ m}^3\cdot\text{h}^{-1}$ and for a minimum flow of $Q_{\min} = 0.7 \text{ m}^3\cdot\text{h}^{-1}$. The fourth scenario simulated the effect of flow-proportional sampling versus time-proportional sampling. For this a diurnal variation according to the method by HSG-Group (Langergraber *et al.* 2008) was generated using the model parameter set: $f_{Q,\min} = 0.42$ (form parameter for minimum flow), $t_{\min} = 0.08 \text{ d}$ (time when minimum flow occurs), $f_{Q,\max} = 1.50$ (form parameter for maximum flow), $t_{\max} = 0.42 \text{ d}$ (time when maximum flow occurs), $f_{N,\max} = 1.60$ (ratio of maximum total Kjeldahl nitrogen (TKN) concentration to mean TKN concentration), $f_{\min,U} = 0.10$ (% fraction of minimum urine flow rate to mean urine flow rate) and $\Delta_{tN} = 0.03 \text{ d}$ (shift of TKN minimum and maximum relative to minimum and maximum flow).

RESULTS AND DISCUSSION

Derivation of a composite sampling strategy by means of simulation

Hydraulic modeling for the derivation of a composite-sampling strategy has already been successfully performed in the context of the Anaerobic Digestion Model No. 1 (ADM1) using pulse injection of lithium chloride (Lübken *et al.* 2015). For the present study, a similar hydraulic modeling approach was developed for the Düsseldorf-Süd WWTP, where NaCl was applied as a tracer to substitute a micropollutant. The simulation considered 24-, 48- and

72-hour composite sampling in terms of dry weather inflow ($Q_d = 1.3 \text{ m}^3 \cdot \text{h}^{-1}$), a rain event ($Q_r = 2.6 \text{ m}^3 \cdot \text{h}^{-1}$) and for a minimum flow ($Q_{\min} = 0.7 \text{ m}^3 \cdot \text{h}^{-1}$). Figure 1 shows exemplarily the RTD of a tracer in the effluent of the Düsseldorf-Süd WWTP in the case of a single pulsed input at a Q_d of $1.3 \text{ m}^3 \cdot \text{h}^{-1}$. While the single pulse keeps in the influent for approximately 1 day, the measurable concentration in the effluent extends over several days. The hatched area in Figure 1 is proportional to the length of composite sampling and hence to the recovery rate of the tracer. It becomes obvious that 24-hour composite sampling will only capture a minor part of the tracer in the effluent, whereas 72-hour composite sampling will recover the highest amount.

From Figure 1 it also becomes apparent that composite sampling beyond a period of 72 hours could increase the recovery rate even further. However, this is not recommendable in order to avoid errors from too long and improper storage.

Considering all simulated events, the benefit of 72-hour composite sampling becomes even clearer (Figure 2). Regarding the 72-hour sampling strategy, an average recovery rate of 80.54% has been found for the dry weather scenario (Q_d), while for the rain event (Q_r) the recovery rate was 95.67%, and 57.51% considering the minimum flow rate (Q_{\min}). The 48-hour composite sampling yielded already significantly lower recovery rates. Since the rain event doubles the inflow rate, the tracer was flushed faster into the effluent, hence also for the 48-hour sample the recovery rate was the highest for the Q_r scenario with 85.91% followed by the Q_d scenario with 61.32% and the Q_{\min} scenario at the very end with 37.63%. Due to the worst recovery rates within this present evaluation, a 24-hour composite sampling strategy is not recommendable for monitoring micropollutants in the field, which is also in line with the findings of Majewsky *et al.* (2011). The Q_r scenario again yielded the highest recovery value with 42.68%, while for Q_d and Q_{\min} the results were significantly low with 22.22% and 10.16%.

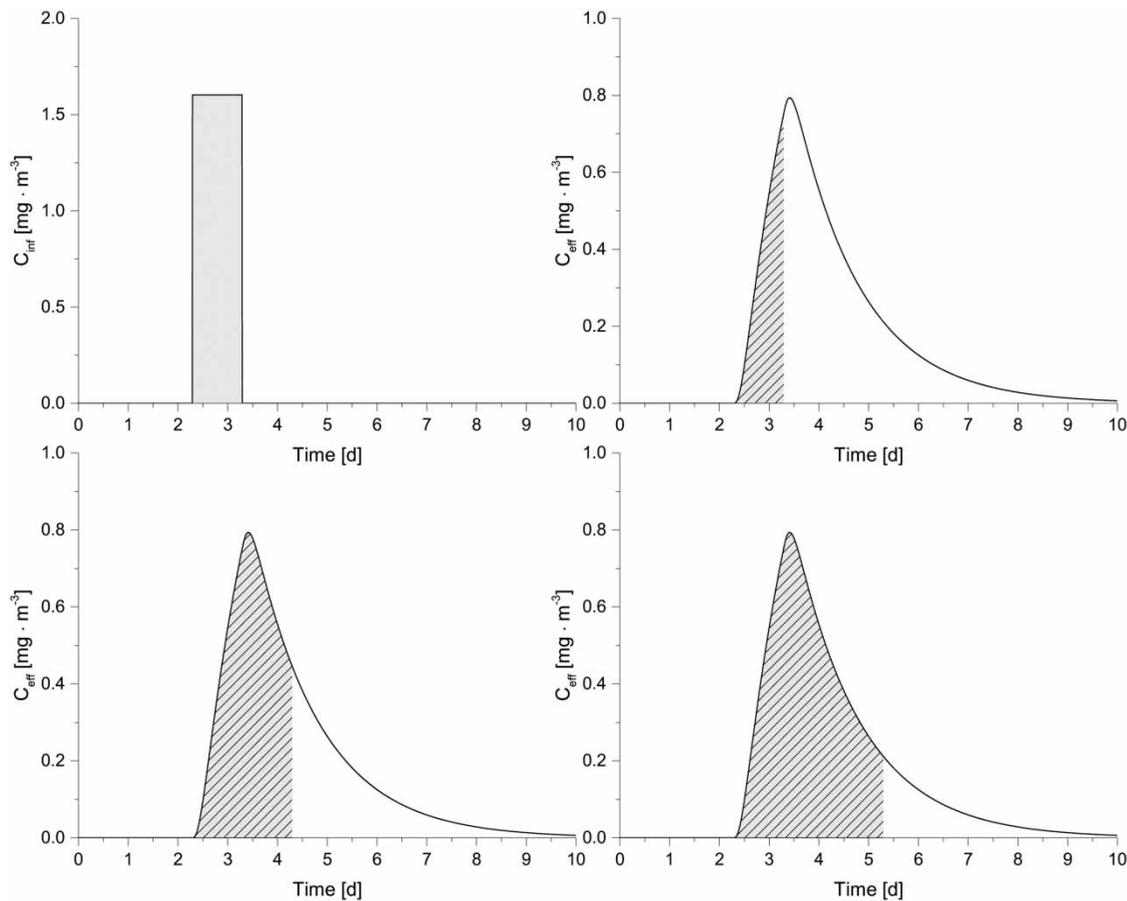


Figure 1 | Exemplary illustration of simulation results from scenario 3 for $Q_d = 1.3 \text{ m}^3 \cdot \text{h}^{-1}$. Concentration of tracer in the influent (C_{infl} , top left), concentration of tracer recovered in the effluent (C_{eff}) after 24-hours (top right), 48-hours (bottom left) and 72-hours (bottom right).

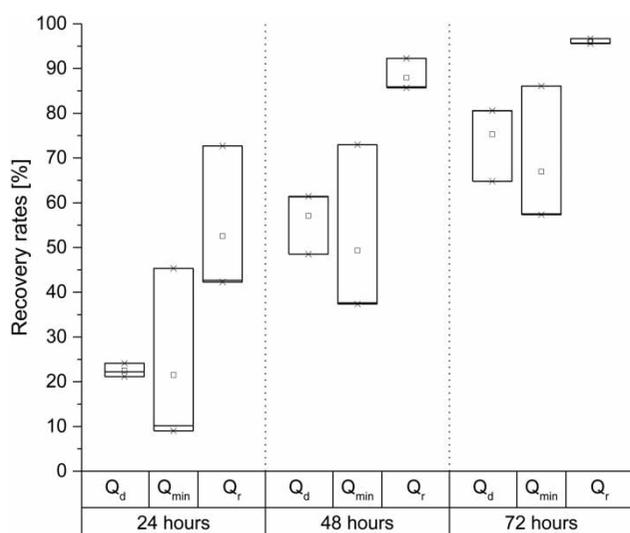


Figure 2 | Threefold determination of tracer recovery rates for the three scenarios comprising dry weather inflow (Q_d), rain event (Q_r) and the minimum flow rate (Q_{min}) for the Düsseldorf WWTP.

It has to be noted that from the perspective of a best sampling strategy a pulse input is much more critical than a continuous input of an inert substance. Knowledge about concentration profiles with high time resolution is highly desirable, but not available in literature. Fast and, above all, cheaper methods for the analysis of micropollutants are still not accessible.

Elimination rates of pharmaceuticals

Based on the previous derived 72-hour composite sampling strategy, simultaneous flow-proportional sampling was performed in the influent and effluent of the semi-technical Düsseldorf-Süd WWTP to monitor the elimination of benzotriazole, carbamazepine, diclofenac, metoprolol and sulfamethoxazole. The first dosage step – $10 \text{ g} \cdot \text{m}^{-3}$ PAC – was started after 90 days of operation without carbon addition to ensure steady state conditions with respect to the biochemical elimination capacity in both lines of the treatment plant. PAC dosing took place in a timeframe of 5 months for each of the two dosage steps. With the direct dosage of PAC into the activated sludge tank the activated sludge is mixed with the added carbon. This leads to a slightly lower proportion of active biomass in the total solids content of the testing line, as the total solids concentration was kept constant at $2.5 \text{ kg} \cdot \text{m}^{-3}$ for both lines. However, no differences have been observed with respect to the elimination of nutrients between both lines. Additionally, a time dependent drift of

measurement results for the analyzed micropollutants since the start of each dosage step was not observed although the amount of carbon incorporated into the activated sludge was increasing over several weeks to reach steady state. After adding to the bulk volume the PAC was rapidly loaded.

The monitored results are summarized in Figure 3 with regard to PAC dosing at rates of $10 \text{ g} \cdot \text{m}^{-3}$ and $20 \text{ g} \cdot \text{m}^{-3}$. The results are completed by reference elimination rates, where no PAC was dosed. For every group depicted in Figure 3 – $10 \text{ g} \cdot \text{m}^{-3}$ PAC, $20 \text{ g} \cdot \text{m}^{-3}$ PAC and reference – 22 samples (11 influent and 11 effluent samples) were analyzed, hence 88 samples per pharmaceutical substance. Sampling regularly took place over the 5 months' operation for each dosage step.

The direct dosing of $10 \text{ g} \text{ PAC} \cdot \text{m}^{-3}$ into the nitrification stage yielded already a significantly enhanced elimination of all of the tested micropollutants, whereas a raised dosage of $20 \text{ g} \text{ PAC} \cdot \text{m}^{-3}$ could not significantly improve or deteriorate the elimination rates, except for sulfamethoxazole. The best elimination rates were found for benzotriazole and metoprolol. At a dosage of $10 \text{ g} \text{ PAC} \cdot \text{m}^{-3}$, benzotriazole was removed by 73% and metoprolol by 67%, while at a dosage

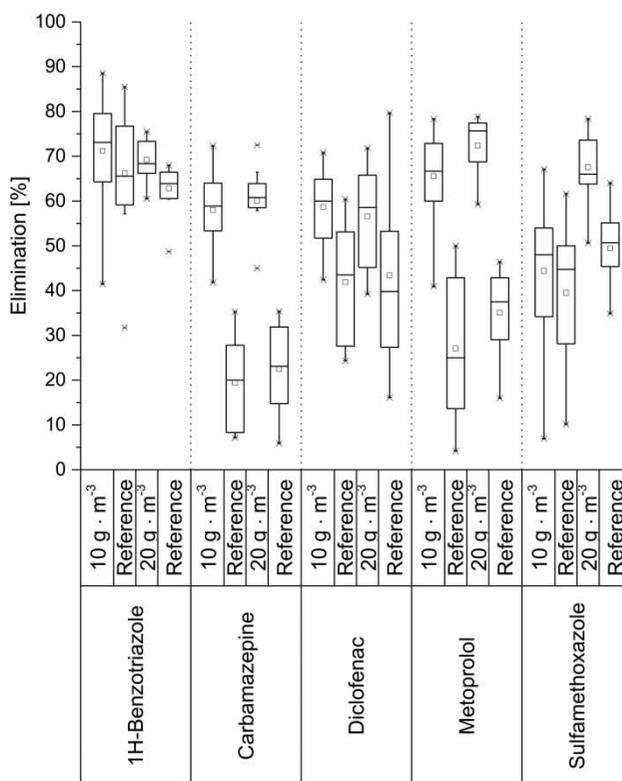


Figure 3 | Elimination rates for benzotriazole, carbamazepine, diclofenac, metoprolol and sulfamethoxazole with regard to $10 \text{ g} \cdot \text{m}^{-3}$ PAC and $20 \text{ g} \cdot \text{m}^{-3}$ PAC.

of 20 g PAC·m⁻³ only the elimination rate for metoprolol was slightly improved to 76%, with benzotriazole removal being 68%. Nevertheless, the obtained elimination rates are in good line with previous studies, although the measurements were performed under different circumstances, such as residence times or reactor designs. Lowenberg *et al.* (2014) achieved a removal efficiency of about 91% for benzotriazole at a dosage of 20 g PAC·m⁻³. For the same micropollutant, removal rates of 38% and 67% were obtained by Altmann *et al.* (2015) at the corresponding PAC dosages. Margot *et al.* (2013) obtained an even higher rate for benzotriazole of 90% at a comparable lower PAC dose of 12 g·m⁻³, whereas the result from Boehler *et al.* (2012) is similar with 95% at 10 g PAC·m⁻³. Comparable elimination rates for metoprolol were obtained in the study by Altmann *et al.* (2015). At this, metoprolol was removed at rates of 46% and 81% at dosages of 10 and 20 g PAC·m⁻³.

Diclofenac and carbamazepine could be both eliminated at a similar range in this study. At 10 g PAC·m⁻³ diclofenac was removed by 60%, and carbamazepine at a rate of 59%. An increased dosage of 20 g PAC·m⁻³ did not improve the elimination rate at all. The obtained rates were 59% for diclofenac and 61% for carbamazepine. Therefore, from a statistical point of view, there was no difference when dosing 10 or 20 g PAC·m⁻³. Nonetheless, carbamazepine, in particular, is known to be the most persistent pharmaceutical that can be removed through conventional wastewater treatment, on average by only 32.7% (Baghdadi *et al.* 2016). The obtained elimination rates are therefore a successful outcome and also in line with other studies. Altmann *et al.* (2015) gained a removal rate of 36% for carbamazepine at 10 g PAC·m⁻³, whereas Margot *et al.* (2013) obtained a 90% removal rate with 12 g PAC·m⁻³. Similar high values were found by Boehler *et al.* (2012) with 78% for 10 g PAC·m⁻³ and 90% for 15 g PAC·m⁻³ and by Mailler *et al.* (2015) with 92% at 10 g PAC·m⁻³. Another comparison can be made with the study by Magdeburg *et al.* (2014), where 50% was achieved for diclofenac and 88% for carbamazepine for 20 g PAC·m⁻³ dose.

Sulfamethoxazole, a moderately adsorptive solute, yielded the lowest elimination rate of 48% at 10 g PAC·m⁻³, but was improved when 20 g PAC·m⁻³ was applied. Here, the median elimination rate was 66%, which is equal to an improvement of +18%. In comparison, Altmann *et al.* (2015) obtained 13 and 32% at 10 and 20 g PAC·m⁻³, which is a similar improvement of +19%. Removal of 28% was obtained in a study by Magdeburg *et al.* (2014) for 20 g PAC·m⁻³ dose, whereas the result obtained by Lowenberg *et al.* (2014) is the closest one to the present study with 56% at 20 g PAC·m⁻³.

Potential practical implementation

The excess carbon used in this study was derived from thermal reactivation of GAC at waterworks, mainly due to abrasion. Approximately 400 tons of this excess carbon, which is considered today only as a waste product, are derived annually from thermal reactivation at the Düsseldorf-Holthausen waterworks. This amount would be sufficient to be applied to the dry weather flow (~26 million m³) of the full-scale Düsseldorf-Süd WWTP over a whole year at a concentration of 10 g PAC·m⁻³. The PAC-AS process as shown in this study is characterized by both low space requirements and monetary investments and hence gives incentives for the implementation of a 'fourth treatment stage' for micropollutant removal.

CONCLUSIONS

In the present study, it could be demonstrated that the use of carbon that is produced through abrasion during the process of regeneration of waterworks carbon can be applied for the elimination of micropollutants during activated sludge treatment. Its applicability was demonstrated in a PAC-AS system for the example of benzotriazole, carbamazepine, diclofenac, metoprolol and sulfamethoxazole, whose removal efficiencies were monitored in the effluent of a semi-technical WWTP. This crosslink constitutes therefore a relatively simple to implement alternative to construction measures regarding advanced treatment units, such as the fourth treatment stage.

The outcomes and advantages can be summarized as follows.

- The addition of PAC into the activated sludge unit enhances the biological stability and improves the effectiveness of the AS system with respect to the removal of micropollutants.
- The results of the present study also emphasize the benefit of dynamic modeling as a tool to derive an optimal sampling strategy for a monitoring survey of micropollutants at WWTP level. The derived length of composite sampling of 72 hours should not be exceeded to avoid errors from too long a sample storage. Flow-proportional sampling should be preferred over time-proportional sampling as the diurnal flow characteristics are considered.
- The yielded removal efficiencies for benzotriazole, carbamazepine, diclofenac, metoprolol and sulfamethoxazole were in good line with published values from the literature, an outcome that can be attributed to the optimized

sampling strategy as well as to the good applicability of reactivated PAC for wastewater treatment.

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