Biologically enhanced granulated activated carbon (BAC) filtration of a mixed industrial/domestic tertiary effluent for reuse

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Abstract Biologically enhanced granular activated carbon (BAC) is a promising wastewater post-treatment to produce a purified effluent suitable for recycle in the textile industry. Three small pilot scale filters were used, filled with three media (two adsorbent media and one non-adsorbent), to compare two different GACs and quantify the biomass contribution to organic removal and colour abatement. A demonstrative scale filter was tested in parallel to evaluate the influence of hydraulic parameters on BAC process efficiency. Biomass contribution was significant when an acclimated biofilm grew on particles; in all cases, wastewater decolorising was due to physical-chemical adsorption only; higher hydraulic load negatively affected the polishing process. Laboratory and semi-industrial scale tests were performed on textile fibres and fabrics using samples of the effluent from the demonstrative filter. The tests confirmed the suitability of the treated water for textile reuse.

Keywords Biological activated carbon; reuse; tertiary treatment; textile wastewater

Introduction Extensive use of underground †and lake water by numerous textile factories in the Lake Como area should be reduced to optimise the use of local water resources. Several treatment schemes have been tested within an EC funded contract (ENV4-CT95-0064, 1999) to improve the quality of tertiary effluent for reuse into textile industries in the Lake Como area (Bergna et al., 1999; Rozzi et al., 1997, 1999). Adsorption by granular activated carbon (GAC) was found to be among the less expensive solutions. A still more efficient treatment can be obtained when biodegradation adds to physical-chemical adsorption on GAC. Biodegradation is carried out by biomass which spontaneously grows on the particles (BAC: Biological Activated Carbon). Its main drawback is an increase of filter clogging that requires more frequent backwashings (Weber et al., 1978; Servais et al., 1992; Dussert and Van Stone, 1994; Scholz and Martin, 1997). In the BAC process, biomass can degrade both dissolved and previously adsorbed compounds, making possible an extension of GAC lifetime (Rice and Robson, 1982; Chudyk and Snoeyink, 1984; Speitel and DiGiano, 1987; Gerald et al., 1989; Olmstead and Weber, 1991; Suzuki, 1997). The BAC process has been extensively investigated to produce drinking water from polluted surface sources, but there are only a few applications in wastewater post-treatment.

Materials and methods The research was carried out at the Alto Seveso wastewater treatment plant (WWTP) located near Como, fed on industrial (mostly textile, about 60% influent COD load) and domestic wastewater. It consists in pre-denitrification/nitrification, coagulation/flocculation, plate settling and ozonation. Four GAC filters were used in parallel: three at small-pilot scale (P1, P2 and P3) and one at demonstrative scale (D). They were fed downflow at constant flowrate using effluent from the ozonation step of the Alto Seveso WWTP. Filters P1 to P3 were made in PVC, while filter D was a stainless steel column; the latter filter was
equipped with 6 water sampling ports through the GAC bed. The main geometrical and hydraulic parameters of the filters are reported in Table 1. Filter P1 was filled with NORIT PK 1–3 GAC (NORIT, Italy); filters P2 and D with PICABIOL GAC (Pica Italia S.p.A., Italy) and filter P3 with anthracite. NORIT PK 1–3 carbon is a peat based steam GAC while PICABIOL is a wood based GAC, activated by phosphoric acid. The main characteristics of the two GACs are reported in Table 2.

TSS, COD, pH and colour were daily analysed on influent and effluent to each filter; pH was measured by a laboratory pH-meter (BE105, Bicasa, Italy); colour was measured as absorbance at 426 nm, 558 nm and 660 nm (optical path of 1 cm) by a spectrophotometer (Shimadzu UV 1204). The three wavelengths used to monitor wastewater colour are usually adopted at the WWTP for the daily controls. NH$_4^+$, NO$_3^-$, NO$_2^-$, TKN, P$_{tot}$ and BOD$_5$ were also analysed weekly on influent and effluent to filter D. Analyses were performed according to Italian Standard Methods (IRSA, 1994), very similar to American Standard Methods (APHA, 1995), on average daily samples, collected and stored at 4°C by 5 automatic samplers (ASP-Station D2, Endress and Hauser, Italy). DO concentration profile through the GAC bed of filter D was measured weekly, for 5 months after one month from the start-up, by a portable DO-meter (HI9142, Hanna Instruments, Italy).

**Adsorption isotherms determination**

Isotherms on the three media and the ozonated effluent (pH: 7.2; soluble (i.e. filtered at 0.45 µm) COD: 55 mg/L; absorbance at 426 nm: 0.020) were performed at 21°C. The media were sieved between 230 and 400 mesh and dried at 105°C before use in a jar-test apparatus (effluent volume in each beaker: 500 mL). After the contact time, residual soluble COD and absorbance at 426 nm were measured on each sample, both in kinetic and isotherm tests. Kinetics of adsorption were determined first to fix the equilibrium time: 240 min. To determine adsorption isotherms, various amounts of the three media were used: from 10 mg to 1,000 mg for NORIT PK 1–3 GAC and from 10 mg to 750 mg for PICABIOL GAC and anthracite.

**Textile tests**

Textile tests were performed using filter D effluent at laboratory and semi-industrial scale, by an independent laboratory and by some textile mills located in Como district. Alkalinity,

| Table 1 | Main geometrical and hydraulic parameters of the GAC filters |
|-----------------|-----------------|-----------------|-----------------|
|                | Unit | P     | D     |
| GAC volume     | m$^3$ | 0.014 | 20.8  |
| GAC height     | m    | 0.8   | 3.15  |
| Flowrate       | m$^3$/h | 0.030 | 40    |
| EBCT (*)       | min  | 28.3  | 31.2  |
| Hydraulic load | m/h  | 1.70  | 6.06  |

(*) Empty bed contact time

| Table 2 | Main characteristics of NORIT PK 1–3 and PICABIOL GAC (NORIT and PICA data sheets) |
|-----------------|-----------------|-----------------|
|                 | NORIT PK 1–3 | PICABIOL |
| Porosity        | cm$^3$/g      | 1.2          | 2.7          |
| Molasses n.     | –              | 500          | 150          |
| Density (*)     | g/L            | 350          | 394          |
| Iodine number   | mg/g           | 800          | 1,000        |
| Ash content     | %              | 7            | 4            |

(*) Directly measured
hardness, conductivity, aldehydes, Al, Cl, surfactants (MBAS, TAS) and some heavy metals (Fe, Cu, Zn, Cr(VI), Cr, Mn) were analysed on the water samples.

Results and discussion

Adsorption isotherms

In Figure 1 the isotherms for NORIT PK 1–3 and PICABIOL GACs are shown for COD and absorbance at 426 nm. Data are well fitted by the Freundlich equation \( \frac{X}{M} = K \cdot C^{1/n} \): parameters estimated by a least square regression, the correlation coefficients and the fitted lines are shown in Figure 1. PICABIOL seems to have better affinity towards the wastewater than NORIT PK 1–3. Adsorption capacity of NORIT PK 1–3 related to COD is more influenced by equilibrium concentration, since the isotherm slope is high, which is a disadvantage in actual operation because of the high variability of wastewater concentration. The adsorption capacities of the two GACs, at the average influent COD concentration during the operating period, are comparable, while the colour adsorption capacity seems to be slightly higher for PICABIOL. The calculated COD removal efficiencies (76% max. for PICABIOL and 55% for NORIT PK 1–3) indicate a significant non-adsorbable organic fraction. Neither COD removal nor absorbance decrease was observed for anthracite during kinetic and isotherm tests. It follows that anthracite can be considered a non-adsorbent media and, therefore, organics removal in filter P3 could only be caused by biodegradation.

Filtration tests

The pilot filters P1 to P3 ran continuously for 170 days and filter D for 231 days. The water volumes treated per unit weight of media by filters P1, P2, P3 and D during the operating period were respectively 23.5, 20.5, 7.4 and 27.2 m³/kg of media. The DO concentration in the ozonated influent to the filters was over the saturation level with respect to air (up to 18 mg/L DO), because the ozone dosed in the tertiary treatment (10–15 mg O₃/L) is produced by liquid oxygen, and promoted aerobic biomass growth on carbon surface; therefore these filters could be considered as biological activated GAC filters (BAC). The wastewater temperature was 15.6 ± 1.4°C: it was constant through the GAC bed of filter D.

In Table 3 TSS, COD and absorbances of the influent to the filters are reported. The influent organic load, both as COD and colour, was very low because of the great dilution induced by heavy and frequent rainfalls during the investigation period. As a consequence, operational conditions were not optimal, since both adsorption rate and biodegradation kinetics were limited by low organic concentration, as showed in Figure 2 where the COD specific removal rate is reported vs. influent organic load for filters P1 and P2. One may

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**Figure 1** Isotherms on NORIT PK 1–3 and PICABIOL: a) COD (NORIT PK 1–3: \( K = 1.42 \times 10^{-4}; 1/n = 3.77; R^2 = 0.962 \); PICABIOL: \( K = 0.36; 1/n = 1.70; R^2 = 0.948 \)); b) absorbance at 426 nm (NORIT PK 1–3: \( K = 8.72; 1/n = 0.96; R^2 = 0.974 \); PICABIOL: \( K = 5.58; 1/n = 0.75; R^2 = 0.979 \))

note actual positive removal values only for organic loads above 2.8 g COD/day/L GAC; for lower values, the COD specific removal rate is negligible and desorption can occur.

Small scale pilot filters

The number of backwashes of filter P3 was 42% higher than the one of filters P1 and P2, possibly due to increased clogging in filter P3 by biomass growth. Backwash stresses induced the partial pulverisation of the media, especially of anthracite and NORIT PK 1–3; PICABIOL appears to be mechanically very resistant, which is important when frequent backwashes are required because of clogging due to biomass growth on carbon particles.

Effluent characteristics of filters P1 to P3 are reported in Table 3. Effluent absorbance at 660 nm was comparable with the detection limit of the spectrophotometer used (0.002), for the three filters, and therefore cannot be considered a significant parameter. The adsorption media were not able to smooth influent variability in the specific operational conditions, possibly due to the great dilution of wastewater, which caused an alternation of adsorption and desorption phenomena, some of significant extent, as one may observe in Figures 2 and 3; in the latter figure the breakthrough COD curve (effluent COD over influent COD vs. Bed Volume (BV) filtered, where BV is the ratio between filtered volume and GAC volume) is plotted for filter P2. A moving average on 7 days data (continuous line) is also plotted, to reduce the very high scattering. The first value in Figure 3 is 0.54 (for filter P1: 0.53), implying that about 55% of the influent COD was not removed by the virgin GAC. This observation possibly implies that dilution limited adsorption rate even at the start-up of the filters, at the maximum adsorption capacity. For filter P3, the breakthrough curve start already at value 1, indicating that no COD removal occurred in the first part of the operating period (about 2 months), when biomass had not yet colonised anthracite.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>P</th>
<th>D</th>
<th>P1</th>
<th>P2</th>
<th>Effluent</th>
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</thead>
<tbody>
<tr>
<td>COD mg/L</td>
<td></td>
<td>53±19</td>
<td>54±19</td>
<td>45±15</td>
<td>42±16</td>
<td>48±18</td>
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<tr>
<td>TSS mg/L</td>
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<td>12±8</td>
<td>3±2</td>
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<tr>
<td>A426 (*)</td>
<td>10^-3</td>
<td>26±12</td>
<td>26±13</td>
<td>17±8</td>
<td>15±8</td>
<td>23±11</td>
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<tr>
<td>A558 (*)</td>
<td>10^-3</td>
<td>13±7</td>
<td>13±4</td>
<td>8±5</td>
<td>8±5</td>
<td>11±7</td>
</tr>
<tr>
<td>A660 (*)</td>
<td>10^-3</td>
<td>6±3</td>
<td>6±4</td>
<td>3±2</td>
<td>3±2</td>
<td>4±3</td>
</tr>
</tbody>
</table>

(*) Absorbance at 426 nm, 558 nm and 660 nm; optical path: 1 cm

Figure 2 COD specific removal rate vs. influent organic load for filters P1 and P2
particles. For BV > 4,000, a slight decrease of the effluent COD over influent COD values was observed due to biomass activity.

A direct comparison of the performance of the three pilot filters, based on COD removal, is shown in Figure 4, where the removed COD load is plotted vs. the influent COD load. The amounts of COD removed by filters P1, P2 and P3 are respectively 55, 74 and 29 g COD per litre of media. In filter P3, adsorption is negligible while biodegradation is appreciable only after the growth of a biofilm on the anthracite particles (BV > 4,000) and the removed COD increases with the influent load. The slope and correlation coefficient of the data (least square linear regression) are reported in Table 4. Three main phases can be observed for filter P1: till BV = 1,800 COD was high (slope of the fitting line: 0.228 ± 0.007 g CODrem/g CODin; $R^2$: 0.977), then it levelled till BV = 4,000 and during the final phase COD removal was again appreciable, but lower than the first phase (data reported in Table 4). For filter P2, COD removal rate (slope of the data) remained always significant. For BV < 1,800, the slope was 0.192 ± 0.010 g CODrem/g CODin ($R^2$: 0.930). Comparable values are found for BV ranging from 1,800 to 4,000. The slope for BV > 4,000 is reported in Table 4. It is worth noting that the COD removal was higher for P1 than P2 during the first phase (BV < 1,800) while the reverse was true for BV > 4,000. The former results contradict isotherm data, which indicated a higher affinity of PICABIOL towards the dissolved COD in the influent. From the observation of Figure 4 one may conclude that the performance of PICABIOL was slightly better than NORIT PK 1–3, even in operational conditions far from the optimal ones. The higher COD removal rate observed during the first phase for filters P1 and P2 are prevalingly due to adsorption; later on, the biofilm building up on GAC particles probably hindered diffusion of organic molecules to the adsorption sites while adsorption capacity was still high, as confirmed by the low amount of COD removed, ranging from 29 to 74 g COD/L, well below data obtained in previous research (Bergna et al., 1999; Rozzi et al., 1997).

In Figure 5 the breakthrough curve for colour (ratio between effluent and influent absorbance at 426 nm vs. BVs filtered) is reported for filter P1. Initial values are of the order of 0.2, suggesting that molecules which contain red chromophores are more adsorbable than most COD (see Figure 4), thus high colour abatement efficiency could be

![Figure 3](https://iwaponline.com/ws/article-pdf/3/3/17/407533/17.pdf)

**Figure 3** Breakthrough curve for filter P2. Continuous line: moving average over 7 days

<table>
<thead>
<tr>
<th>Slope ($^*$)</th>
<th>P1</th>
<th>P2</th>
<th>P3</th>
<th>D</th>
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<tr>
<td>$R^2$</td>
<td>0.960</td>
<td>0.987</td>
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</table>

$^*$ The slope coefficient is expressed in g COD removed per g COD influent load.

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**Table 4** Slopes and correlation coefficients of the linear regression on data in Figure 4 after 4,000 BV filtered (average value ± standard deviation), for filters P1, P2, P3 and D.
reached with virgin carbon, according to the isotherm for NORIT PK 1–3 (max. calculated efficiency: 96%). The same considerations apply to PICABIOL. Abatement efficiency rapidly decreased for filters P1 and P2 until 790 BV filtered, after which, two abatement levels can be observed before and after 4,000 BV filtered: 0.56 ± 0.10 and 0.75 ± 0.15 respectively for P1 and 0.53 ± 0.09 and 0.69 ± 0.16 for P2. The scattering of the absorbance data is high, but PICABIOL seems to have a better performance than NORIT PK 1–3 also in this case. Data for filter P3 are uniformly dispersed around 1 (0.91 ± 0.24) which indicates that biodegradation alone is unable to abate colour. The same results have been obtained for absorbance at 558 nm (not reported).

**Demonstrative scale filter**

To evidence aerobic biological activity, the DO concentration profile through the GAC bed of filter D was measured; in Figure 6, DO concentrations are plotted vs. filter depth. DO decreased through the GAC bed according to a constant rate (3.6 mg DO consumed per m of bed): in Figure 6 the regression line and the correlation coefficient are reported.

In Table 3 the main parameters of the effluent to filter D are reported. The COD removed is plotted vs. influent COD load in Figure 4 (filter D removed 100 g COD/L GAC). Data for filter D and P2 are comparable only for BV filtered < 4,000, when adsorption was the prevailing removal mechanism; later on, data are well distributed on a straight line which has a
slope lower than the one related to P2 (Table 4). Similar qualitative considerations apply to colour abatement for P2 and D: after a rapid decrease of absorbance, data (ratio between effluent and influent absorbance at 426 nm) appear to be uniformly distributed around an average value (0.80 ± 0.16), lower than the one calculated for P2. Therefore, in the given operational conditions, the higher hydraulic load for filter D seems to negatively affect COD removal and colour abatement, probably because it influences diffusion and adsorption rates of organic molecules into the biofilm and the GAC particles.

Textile tests
Samples of D effluent, after partial softening, and samples of the water supplied by textile industries of the Como district, were used to perform textile tests (scouring and dyeing) at laboratory and semi-industrial scale on fabrics (silk, viscose, acetate/viscose, polyester and wool/silk) and silk yarns, according to the same dyeing recipe. Acid, reactive, direct, dispersed dyes have been used at colour densities considered critical for the yarns and fabrics processed. Two indices were evaluated: colorimetric co-ordinates and resistance to scouring on dry and wet textiles. Results were fully acceptable and therefore the effluent of filter D can be considered suitable for textile reuse.

Conclusions
Biomass contributed to COD removal (about 0.1 g of COD removed per g of influent COD) after a stable biofilm colonised the media. Dyestuff molecules and other organics appear to have a different removal and therefore biodegradation can contribute significantly to COD removal but not to colour abatement. PICABIOL, with respect to NORIT PK 1–3, had a more constant performance, especially for COD removal, probably because its structure enhances biomass growth, determining a synergetic effect. Moreover it showed a greater mechanical resistance to pulverisation due to backwashings. A higher hydraulic load (1.7 m/h to 6.1 m/h) negatively influenced BAC process efficiency, with a 22% decrease in organics removal. Textile tests confirmed the suitability of the BAC effluent for reuse within textile processes. The obtained results are promising, but additional research in more representative operating conditions is necessary to better understand the BAC process.

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