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Effects of water quality changes on performance of biological activated carbon (BAC) filtration

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ABSTRACT

Biological activated carbon (BAC) filtration is an important treatment step in the production of drinking water especially if drinking water is produced from surface water. The performance and processes within a BAC filter have been of interest for researchers since the 1980's, mainly because of its ability to remove natural organic matter known as disinfection precursors. A malfunction of one of the pre-treatment steps might affect the feed water quality into the BAC filters. The main objective of this study was to determine the immediate response of the BAC filters to a rapid change in feed water quality. It was shown that with the studied setup it was possible to compare the effect of different pre-treatment steps and subsequent different water qualities on the performance of the BAC filters on the long term adaptation. However, especially the immediate response was not studied in detail before. All filters were able to mitigate a sudden change in feed water quality, either through improved adsorption or increased activity of the biomass on the filter. As a result of this resilience against sudden changes, it is therefore concluded that there is no direct need for very stringent on-line monitoring and continuous adjustments of the feed water quality of the BAC filters. The addition of phosphate resulted in the lowest dissolved organic carbon (DOC) concentration in the effluent of the BAC filters. In this study the influence of intact cells in the feed water on the performance of the BAC filters was shown to be limited.

1. Introduction

Granular activated carbon (GAC) filters in drinking water treatment are used for the removal of organic compounds such as organic micro-pollutants, organic matter causing color, taste and odor and precursors for disinfection by-products [1]. Removal of these organic compounds takes place by means of adsorption. The presence of natural organic matter (NOM) in the water can have a negative impact on the organic micro-pollutant removal due to pore blocking and competition [2,3]. When GAC filters are preceded by an oxidation step, the large organic molecules are converted into low molecular weight molecules enhancing the biological activity in the water and on the GAC grains. This turns the GAC filters into biological activated carbon (BAC) filters [4,5], where simultaneous removal of NOM by adsorption and biodegradation takes place. This has a positive effect on the required regeneration frequency, due to less pore blocking by NOM, and typically makes BAC filtration more (cost) effective than GAC filtration [4]. The drawback of converting NOM into biodegradable NOM is the negative effect this has on the biological stability of the drinking water [6]. A balance exists between the amount of biodegradable NOM that is produced during oxidation and the amount that can be biodegraded in the BAC filters [7].

Biodegradation of NOM takes place via micro-organisms (biomass) that grow on the external surface and in the macro-pores of the BAC filter grains [8]. The biomass activity determines the degradation rate of NOM [9]. Persson et al. [10] described that the activity and concentration of biomass depends on the concentration of nutrients, intact cell concentration in the feed water and predation. In 2006, Van der Aa et al. set up a computer model to describe the simultaneous adsorption and biodegradation of NOM in BAC filters. From this model it was concluded that, in addition to substrate concentration and predation, maintenance, attachment/detachment and transport of bacteria could not be neglected when determining the biomass development. This is in line with what Uhl and Gimbel [11] found, indicating the need for deposition of bacterial cells from the influent to maintain a solid biofilm. On the other hand Simpson [12] described that efficient substrate removal took place even when the number of bacterial cells in the influent was low. Recently, Liao et al. [13] found that nutrients play a

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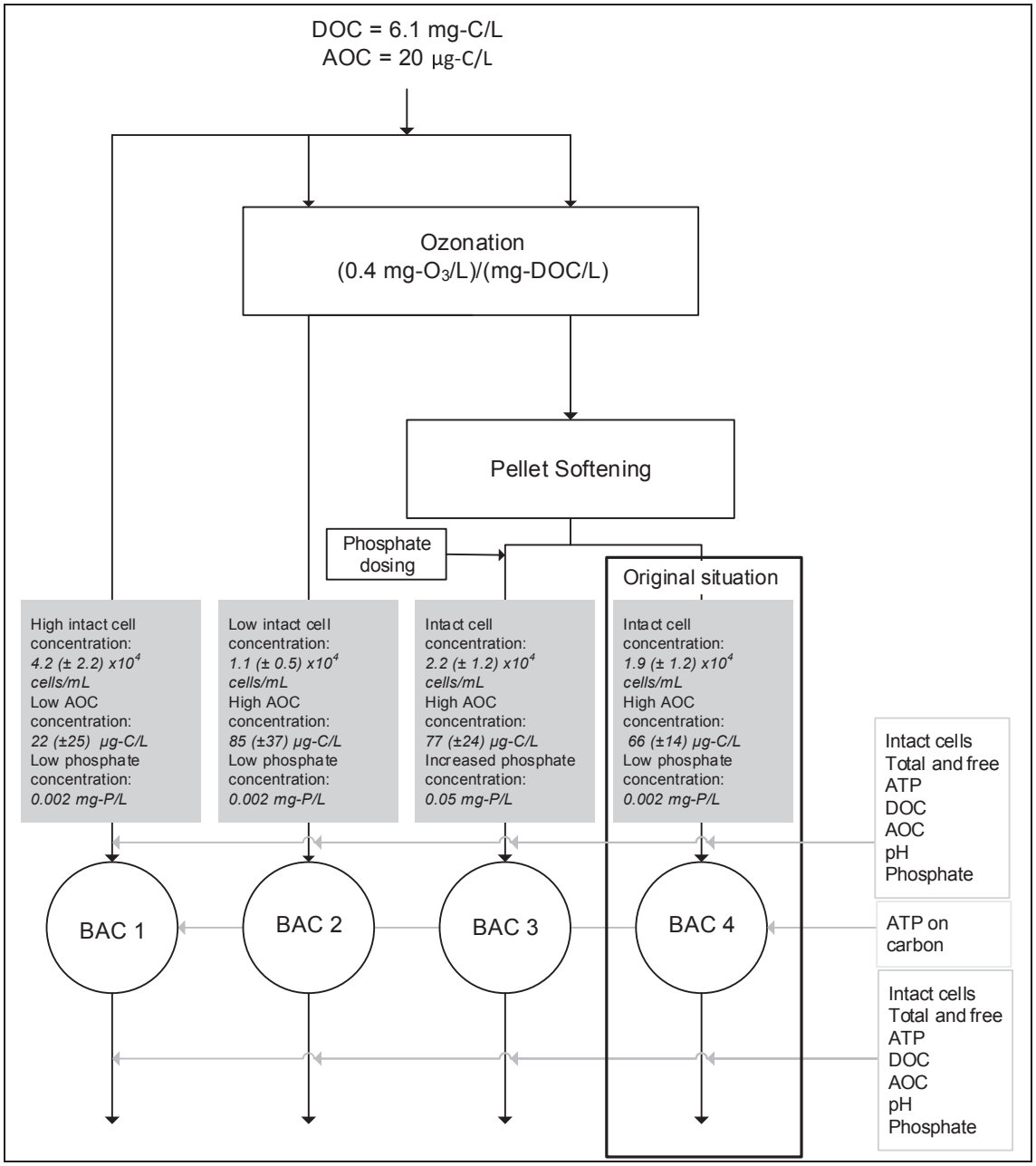

Fig. 1. Schematic overview of pilot plant BAC filters.

more important role than bacterial cells in the development of biomass density. However, when the nutrient conditions are similar, the biomass in the influent becomes the determining factor.

Due to the complexity of the measurements to determine bacterial cells and biomass activity, there has been a lack of available datasets representing the biomass development over a long period of time and over the height of the filter bed [14]. In 2005, Hammes and Egli [15] published a method for the determination of intact cell concentrations in water with flow cytometry. It was shown that this technique was a rapid, easy and sensitive method to determine concentrations of intact cells in water [16]. By combining flow cytometry data with adenosine tri-phoshate (ATP) measurements it was possible to describe the initial colonization and development of biomass [14]. Furthermore it was investigated if a correlation existed between biomass growth and the assimilable organic carbon (AOC) concentration. AOC represents the readily available carbon for cell growth. No correlation was found between the AOC concentration and biomass growth.

Micro-organisms require carbon (C), nitrogen (N) and phosphorus (P) as main nutrients. Van der Kooij et al. [17] established that the typical ratio of C:N:P is 100:10:1. Therefore it is often assumed that biodegradable organic matter is the growth limiting nutrient for NOM reducing bacteria. However, in some cases it was found that microbial growth in drinking water was limited by phosphorus [18–21] or nitrogen [13] instead of carbon. Dhawan et al. [22] investigated the influence of nutrient supplementation on DOC removal by BAC filters by testing different types of ratios. Increased nutrient concentrations beyond the typical ratio of 100:10:1, resulted in increased removal of DOC. In addition to DOC removal, the effect of increased nutrient concentrations on biomass development was determined by means of phospholipid and ATP analysis. In this case no correlation could be found between the DOC removal and the biomass measurements applied.

Although researchers have tried to find correlations between biomass development and the observed biodegradation in the BAC filters

under varying operating conditions, no clear correlations have been found [14,22]. This underlines the complexity of the processes taking place in the BAC filters. BAC filters are frequently used in the production of drinking water for the removal of organic micro pollutants and organic matter, especially when produced from (humic rich) surface water. Differences in feed water quality are the result of pre-treatment steps (coagulation/flocculation, ozonation and phosphate addition), commonly applied in the production of drinking water. These long term effects of changes in feed water quality were investigated and compared to previously published research. However, to the best of our knowledge, the immediate responses of BAC filters, which have been in operation for multiple years, to rapid changes in feed water quality have not been studied previously. A malfunction of one of the pre-treatment steps might affect the feed water quality into the BAC filters, therefore being representative for possible events taking place in common drinking water treatment trains. Investigating the immediate responses of BAC filters to changes in water quality is an important step towards understanding the ability of BAC filters to produce a stable drinking water quality under varying conditions and was the main objective of the presented study.

2. Materials & methods

2.1. Pilot plant set-up and operation

The experiments were carried out at the pilot plant of Waternet, the water-cycle company of Amsterdam and surrounding areas, location Weesperkarspel (the Netherlands). The pilot plant was fed with humic rich surface water (average DOC concentration of 9.2 mg-C/L), pre-treated by coagulation/sedimentation, followed by 100 days retention in a reservoir and subsequently filtered over rapid sand filters. The pilot plant includes all treatment processes of the full-scale plant, consisting of ozonation, pellet softening, BAC filtration and slow sand filtration as final disinfection step.

Four BAC filters with a surface area of 0.43 m^2, bed height of 3.2 m, filtration velocity of 4 m/h and an empty bed contact time (EBCT) of 48 min, were operated at the pilot plant. Before the experiment started, the pilot plant was operating at the same setpoints as the full-scale plant. The filters were fed with pre-ozonated (0.42 mg-O_3/L)/(mg-DOC/L) and softened water, resulting in a feed water quality with an average DOC concentration of about 5.5 mg-C/L and an AOC concentration of about 60 µg-C/L. The activated carbon (type: Norit GAC 830P) was taken from the full scale plant and had been in service for 5 years in the pilot plant to minimize the effect of adsorption during the experiments. Adsorption processes were neglected to represent the worst case scenario from an operating point of view. The BAC filters are supposed to be less robust if only biodegradation processes are considered, than if both biodegradation and adsorption takes place.

At the start of the experiment (day 1), the feed water quality to BAC 1, 2 and 3 was changed (see Fig. 1), while the feed water quality of BAC 4 remained unchanged. BAC 4 served as a reference filter containing a moderate concentration of bacterial cells due to bacterial growth in the softening step, and high biodegradable NOM concentration, measured as AOC. BAC 1 was fed with pre-treated water only (coagulation/sedimentation followed by rapid sand filtration). This resulted in high intact cell concentrations and a low AOC concentration. BAC 2 received ozonated water directly, resulting in the highest AOC concentration and low concentration of intact cells. BAC 3 was fed with water similar to the original situation (moderate concentration of intact cells and AOC) with added phosphate in the form of phosphoric acid (approximately 50 µg-P/L) to promote bacterial growth in the BAC filter. Phosphate was added to be able to investigate the effect of coagulation/sedimentation on biological activity in the filter bed.

2.2. Sampling and water quality analysis

Water samples were taken from the influent and effluent of the four filters. The natural organic matter, dissolved in water, was determined by measuring the DOC concentration. The DOC, pH and phosphate concentration were analyzed according to the Standard Methods [23]. The readily available carbon for cell growth was determined by measuring the AOC concentration. AOC was measured in duplicate, applying the simultaneous incubation of the strains P17 and NOX as described by Van der Kooij and Hijnen [24].

Vital et al. [25] showed that flow cytometry for measuring cell concentrations and ATP analysis were able to describe major bacteriological changes in the Weesperkaspel full scale treatment plant. In this study similar methods were used, with intact cell concentration being determined by fluorescent staining and flow cytometry as described by Hammes et al. [16]. The total ATP was determined using the BacTiter-Glo™ reagent (Promega Corporation, Madison, WI, USA) and a luminometer (Glomax, Turner Biosystems, Sunnyvale, CA, USA) as described by Hammes et al. [26]. The data were collected as relative light units and converted to ATP by means of a calibration curve made with a known ATP standard (Promega). Free ATP was determined by filtering the sample through a 0.1 µm sterile syringe filter (Millex®-GP, Millipore, Billerica, MA, USA), followed by analysis described above. The cell-bound ATP was calculated by subtracting the free ATP from the total ATP. The intact cell concentration and cell bound ATP enabled the calculation of the ATP-per-cell concentration, as expressed in Eq. (2).

Activated carbon samples were taken from 0.1 m and 1.6 m below the top of the filter bed, corresponding with an EBCT of 1 and 22 min, respectively. The biomass activity of the carbon samples was determined by ATP on carbon according to the method described by Magic-Knezev and van der Kooij [27].

2.3. Calculations and statistics

To compare the microbiological activity between the four BAC filters, the overall ATP concentration in the filters was determined as described by Velten et al. [14]. Each filter was partitioned into two parts. The line of demarcation was in the middle of the two sampling points. In line with Velten et al. [14] it was assumed that the measured ATP on carbon at the sampling points represented the average ATP concentration on carbon for the segment. This resulted in the following equation for the determination of ATP in filters:

$$\begin{aligned} ATP_{segment1} &= A \cdot h_{layer1} \cdot \overline{ATP_{on\_carbon\_EBCT\_1\,min}} \cdot \rho_{NoritGAC830P} \\ ATP_{segment2} &= A \cdot h_{layer2} \cdot \overline{ATP_{on\_carbon\_EBCT\_22\,min}} \cdot \rho_{NoritGAC830P} \\ ATP_{overall} &= ATP_{segment1} + ATP_{segment2} \end{aligned} \quad (1)$$

where A represents the surface area in m^2, h_{layer1} represents the height of layer 1 in m, $ATP_{on\_carbon\_EBCT\_1min}$ represents the ATP concentration on carbon at an EBCT of 1 min in (g ATP/g carbon) and $\rho_{NoritGAC830P}$ represents the density of Norit GAC830P carbon in g/m^3.

The ATP-per-cell was determined by Eq. (2) [28]:

$$ATP - per - cell = \frac{Cell - bound\_ATP \times 10^{-9}}{Intact\_cells \times 10^3} \quad (2)$$

where *Cell-bound ATP* is expressed in ng/L, the *Intact cells* is expressed in cells/mL. The *ATP-per-cell* is expressed in g ATP/cell.

The Mann-Whitney U test (ranksum), a nonparametric test fit in Matlab [29] to test skewed datasets (not normally distributed), was used to evaluate statistically if the influent and effluent water qualities of the BAC filters were significantly different. The statistical difference is expressed by the p-value, where values smaller than 0.05 indicate a significant difference with a confidence interval of 95%. For average values the standard deviation was displayed as well as the number of samples (n) based on which the average was determined.

2.4. Immediate response and long term adaptation

The sampling frequency at the beginning of the experiment was high and taken at day 1, 5, 8 and 13 after switching to the different pre-treatment steps. To determine the effects of the change in pre-treatment on the long term adaptation of the biomass and the biodegradation efficiency, the filters were sampled once every four weeks for a period of 436 days.

3. Results

3.1. Differences in influent quality

Comparison of the influent qualities of BAC 1 and BAC 4 showed the effect of additional treatment steps versus direct treatment of pre-treated surface water (Fig. 1). The pre-treated surface water contained low AOC concentrations since most biodegradable organic matter had been consumed during the long residence times in pre-treatment. Without disinfection by ozonation the intact cell concentration in BAC 1 was high. The feed waters of BAC 1 and 4 were statistically different for both intact cells and AOC (p-values of 0.005 and 0.003, respectively). Comparison of BAC 2 and 4 showed the effect of a limited concentration of intact cells (p-value of 0.02) in the feed water due to the addition of ozone, and subsequent disinfection of bacterial cells. By applying ozone, part of the organic matter was converted into biodegradable organic matter, expressed by the increased AOC concentrations in the influent of BAC 2 [30]. Biological growth, occurring in the pellet softening step, reduced the AOC concentration and increased the concentration of intact cells in the influent of BAC 3 and 4 [31]. No significant statistical difference was observed between the AOC concentrations in the feed of BAC 2, 3 and 4, although BAC 3 and 4 were preceded by the pellet softening step. The addition of phosphate to the feed water of BAC 3 provided insight in possible limitation in phosphate (p-value of 0.0002), when comparing BAC 3 with BAC 4.

3.2. Biomass development

The active biomass on the granular activated carbon grains was measured at the start of the experiment at two different bed heights, equivalent with an EBCT of 1 and 22 min. Based on these measurements the total amount of ATP on the activated carbon in the filters was determined using Eq. (1). At the start of the experiment the total ATP was statistically similar for all four filters (Table 1), with p-values of 1.0, 0.1 and 0.3 when comparing the individual filters with BAC 4. The increase in active biomass calculated for BAC 1 and BAC 4 (reference filter) was limited compared to the increase in ATP measured for BAC 2 and 3.

The highest concentrations of active biomass were found in the top of the filter and decreased with increasing depth (Table 2). Similar to what was found by Urfer and Huck [32]. The measured active biomass concentrations were in the same order of magnitude as found by Van der Aa et al. [2], Velten et al. [14].

3.3. Intact cell concentration, cell-bound ATP and bacterial changes

A comparison was made between changes in intact cells, over the individual filters, immediately after start of the experiment (first three days) and after reaching steady state (average of 8 samples taken during the period of one year), see Table 3. In BAC 1 the removal percentage of intact cells varied from 30% immediately after start to an average 23% removal after reaching steady state. Similarly BAC 4 also showed a small variation with 47% removal of intact cells the first three days after start experiment while based on the yearly average 42% removal was observed. The intact cell concentration in the effluent of BAC 2 showed an increase of 442% compared to the influent of BAC 2 in the first three days after the start of the experiment, indicating a considerable growth or detachment of bacteria. An increase in intact cells was still observed in BAC 2 during the steady state situation, but only with a percentage of 35%, still indicating growth or detachment of bacteria [33]. BAC 3 also responded with a direct increase in intact cells of 28% when changing the feed water quality whilst in the steady state situation a decrease of 26% was observed over the filter.

In the steady state situation, the concentration of intact cells in the feed water of BAC 1 was the highest, due to the absence of a disinfection step, with on average 3.5×10^4 ($\pm$ 1.7×10^4, n = 8) cells/mL, being about two times higher than in the feed of BAC 4 (Fig. 2). The cell-bound ATP concentration in the feed of BAC 1 was 50% higher, thus resulting in lower ATP-per-cell concentrations. The intact cell concentration in the effluent of BAC 1 decreased on average with 23% compared to the influent, while the cell-bound ATP concentration decreased with 28%.

The intact cell concentration in the feed water of BAC 2 was the lowest with on average 9.7×10^3 ($\pm$ 4.7×10^3, n = 8) cells/mL, 43% less than in the feed of BAC 4, as a result of the applied ozone step. However, the cell-bound ATP concentration was similar to that of the feed of BAC 4. This deviation in pattern between intact cell and cell-bound ATP concentration during ozonation has also been observed by Vital et al. [25].

The increased intact cell concentration in the influent of BAC 3 and 4 was the result of biological growth taking place in the pellet softening step, as also observed by Hammes et al. [31] and Vital et al. [25]. The concentration of intact cells in the influent of BAC 3 and 4 was similar with 1.7×10^4 ($\pm$ 0.8×10^4, n = 8) cells/mL. Both filters showed a decrease in intact cells after filtration with 26% for BAC 3 and 42% of BAC 4. The large variation in cell-bound ATP measurements also resulted in the ATP-per-cell concentration to fluctuate a lot, making it difficult to compare the results (data not shown).

Table 1
Total amount of ATP on activated carbon at the start of experiment (average of three measurements) and end of experiment (average measurement based on last three sampling days).

	BAC 1 [g ATP]	BAC 2 [g ATP]	BAC 3 [g ATP]	BAC 4 [g ATP]
Start of experiment	0.24	0.33	0.24	0.24
End of steady state situation	0.28	0.60	0.72	0.33
Change in ATP concentration	0.03	0.27	0.48	0.09

3.4. Phosphate uptake

The phosphoric acid dosage upstream of BAC 3 resulted in significant higher phosphate concentrations in the influent. At the start of the experiment almost all phosphate dosed was taken up completely, measured by the difference between influent and effluent concentration. After 5 days the uptake started to decrease gradually until a steady state uptake of about 0.005 ($\pm$ 0.001, n = 5) mg-P/L was reached (Fig. 3).

The ortho-phosphate concentration in the influent of BAC 3 showed some variation with higher concentrations in the beginning of the experiment. Measurements showed that the concentrations in the influent of BAC 1, 2 and 4 were stable, and variations in BAC 3 was therefore the result of variations in dosing, due to variances in the operation of the dosing pump. Since the uptake was significantly lower than the measured influent concentrations it can be concluded that the fluctuations in the dosing did not interfere with the intended setup of the experiment.

3.5. Removal efficiency DOC

The non-ozonated water (BAC 1 feed) had the highest DOC concentration (6.0 $\pm$ 0.1 mg/L, n = 8) and, consequently, also had a yellowish color (Fig. 4a). The color was subsequently removed by

Table 2
ATP on carbon concentrations in steady state situation based on the average (and standard deviation) of the last three sampling days for different EBCTs.

	BAC 1 g ATP/g carbon	BAC 2 g ATP/g carbon	BAC 3 g ATP/g carbon	BAC 4 g ATP/g carbon
EBCT 1 min	$1.12 (\pm 0.60) \times 10^{-6}$	$2.93 (\pm 1.87) \times 10^{-6}$	$3.38 (\pm 1.35) \times 10^{-6}$	$1.21 (\pm 0.68) \times 10^{-6}$
EBCT 22 min	$0.20 (\pm 0.03) \times 10^{-6}$	$0.26 (\pm 0.11) \times 10^{-6}$	$0.35 (\pm 0.09) \times 10^{-6}$	$0.29 (\pm 0.04) \times 10^{-6}$

Table 3
Difference in intact cells measured in influent and effluent expressed in change over the filter by subtracting the effluent concentration minus the influent concentration divided by the influent concentration and multiplied with 100 to obtain the percentage.

	Immediate response [effluent – influent] expressed in %	Steady state [effluent – influent] expressed in %
BAC 1	−30	−23
BAC 2	+442	+35
BAC 3	+28	−26
BAC 4	−47	−42

treatment with ozone which slightly reduced (not significantly) the DOC concentration and significantly increased the readily biodegradable NOM concentration (expressed as AOC).

Direct feed of ozonated water (BAC 2) resulted, immediately after the start of the experiment, in a lower DOC effluent concentration with 4.2 (± 0.03, n = 2) mg/L and higher removal percentage of 35% versus 24% and 23% for BAC 3 and 4, respectively. After reaching steady state, the removal percentages in BAC 2 were still the highest with 24%, followed by BAC 3 with 22% and BAC 4 with 21% (Fig. 4b). The lowest DOC concentrations was reached by BAC 3 with 4.5 (± 0.2, n = 8) mg-C/L (not significant with p-value of 0.1679) followed by BAC 4 with 4.7 (± 0.2, n = 8) and BAC 2 with 4.8 (± 0.2, n = 8) mg-C/L (not significant with p-value of 0.0891).

3.6. Bacterial growth potential produced water

The measured AOC concentration in the effluent was used as an indicator of the bacterial growth potential of the water. No additional source of nutrients were introduced when measuring the growth of the two bacterial strains *Pseudomonas fluorescens* P17 and *Spirillum* spp. NOX. Therefore it was assumed that the measured AOC concentration represented the actual growth potential of the water instead of just carbon limitation [34]. Thus, the lower the AOC concentration the lower the bacterial growth potential. The effluent from BAC 1 had, in steady state, the lowest AOC concentration. The application of a disinfection step for the treatment of surface water before BAC filtration, has shown to be beneficial with respect to regrowth potential and control of disinfection by-products, such as chlorinated hydrocarbons [4]. Because of this, BAC 1 was not discussed in further detail when evaluating the growth potential.

The influent AOC concentration of BAC 2 showed the highest variability in the measurement, caused by seasonal variation of the influent quality and subsequent influence of ozone on production of biodegradable organic matter (Fig. 5a). The influent AOC concentration of BAC 3 and 4 showed less seasonal influences as a result of the pellet softening step between the ozone and BAC filters, levelling out the differences. The AOC concentration in the feed water of BAC 2 was the highest followed by BAC 3 and 4. The difference observed between the AOC concentration in BAC 3 and 4 is explained by the addition of phosphate and the AOC measurement representing the growth potential instead of just the carbon growth potential, indicating phosphate limitation of the water [34].

When comparing the AOC concentration of the produced water from BAC 2, 3 and 4, BAC 2 resulted in the lowest AOC concentration for both the immediate response (17 µg-C/L) as well as the steady state situation (36 ± 6 µg-C/L, n = 4). With the highest AOC concentration, on average 74 (± 24, n = 4) µg-C/L, in the influent of BAC 2, this also resulted in the highest removal percentage of 51% (Fig. 5b). Statistical analysis showed that the removal percentages were not significantly different. The immediate response of BAC 3 resulted in a higher AOC concentration in the effluent than BAC 4, while the steady state

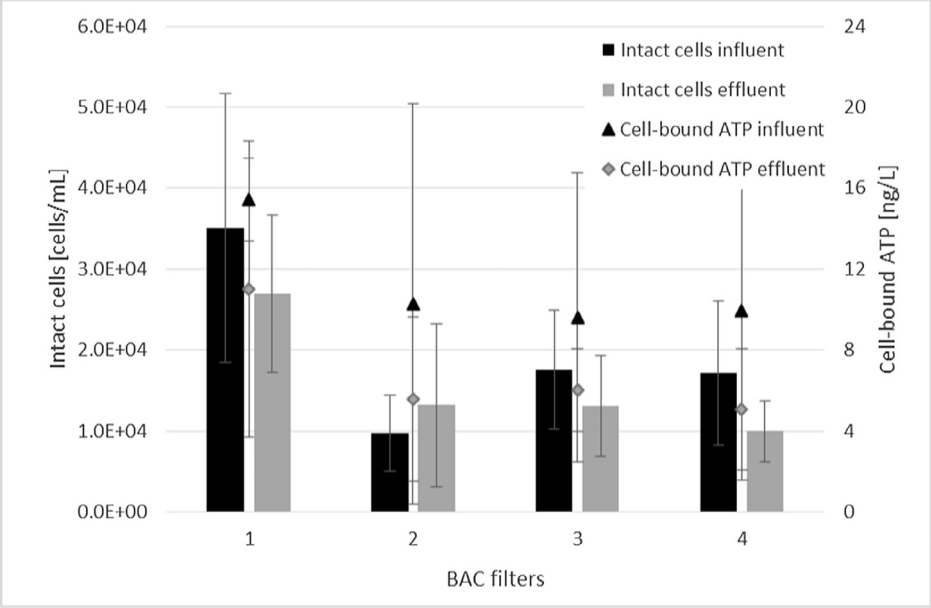

Fig. 2. Steady state (average of 8 measurements, covering a one year timeframe) intact cell concentration (left axis) and cell-bound ATP (right axis) measured in both influent as effluent of the four BAC filters.

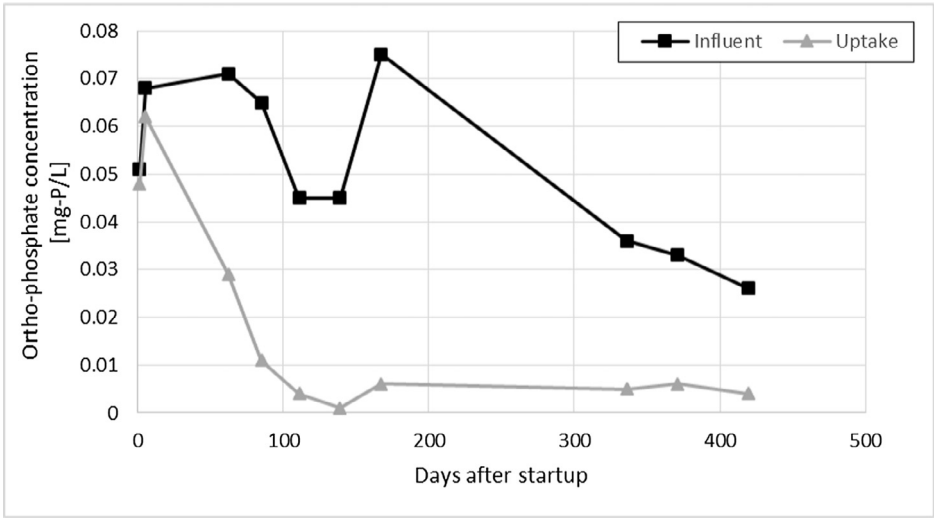

Fig. 3. Measured ortho-phosphate concentration in influent of BAC3 and calculated uptake of ortho-phosphate based on the difference between influent and effluent.

consolidated with removal percentages similar to the reference situation (BAC 4). Even though the AOC concentrations and removal percentages were not statistically different, it was shown that sufficient nutrients positively influence the performance of the BAC filters and bacterial growth potential of the water.

4. Discussion

4.1. Long term effects of changes in treatment plant set-up

The ATP on carbon, intact cell concentration, cell-bound ATP and ATP-per-cell concentration provided insight in the behavior of the filters and development of biomass. All filters showed an increase in active biomass concentration until the steady state situation was reached after 111 days. This was in line with the periods between 2 and 8 months that have been reported for the biomass to fully adapt to the new feed water quality [8,2]. The highest concentrations of active biomass were found in the top of the filter and decreased with increasing depth, similar to what was found by Urfer and Huck [32]. The measured active biomass concentrations were in the same order of magnitude as found by Van der Aa et al. [2], Velten et al. [14].

The ATP-per-cell concentration is correlated with size and physiological activity of the cells [26]. Lower ATP-per-cell concentrations indicate the presence of smaller or less active cells in the feed of BAC 1 compared to the feed of BAC 4. The decrease in intact cells and cell-bound ATP coincided with the limited growth of biomass found in BAC 1, measured through the ATP on carbon concentrations (Tables 1 and 2). Although the intact cell concentration in the feed water of BAC 2 was lower, during steady state, the intact cell concentration in the effluent of BAC 2 was 32% higher than in the effluent of BAC 4 (Fig. 4). The increase in intact cell concentration showed a considerable growth of bacteria in BAC 2. This was also reflected by the ATP on carbon concentrations increasing in time in BAC 2 (Table 1). The increase in ATP on carbon concentration in BAC 3 reflected biological growth, even though the intact cell concentration decreased. For BAC 4 no direct increase in ATP was expected since the filter had been operating under

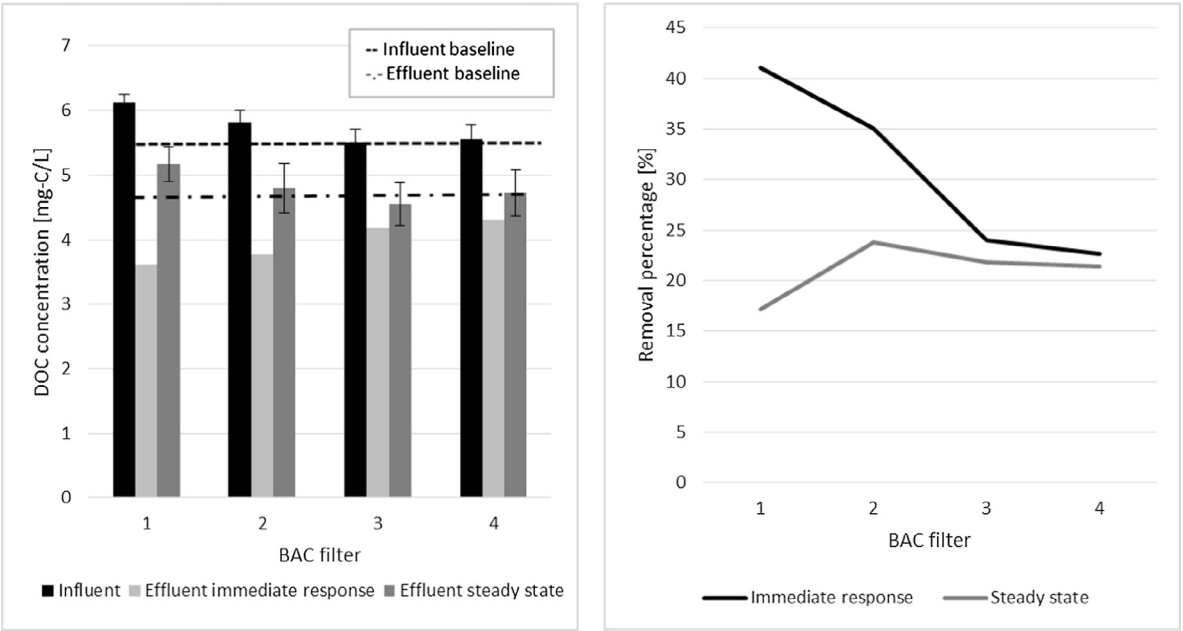

Fig. 4. (a) DOC concentration measured in influent and effluent for both the immediate response and steady state situation (left) and (b) removal percentage DOC per filter for immediate response and steady state situation. The indicated baseline refers to the original situation (BAC 4) and is included for easy comparison.

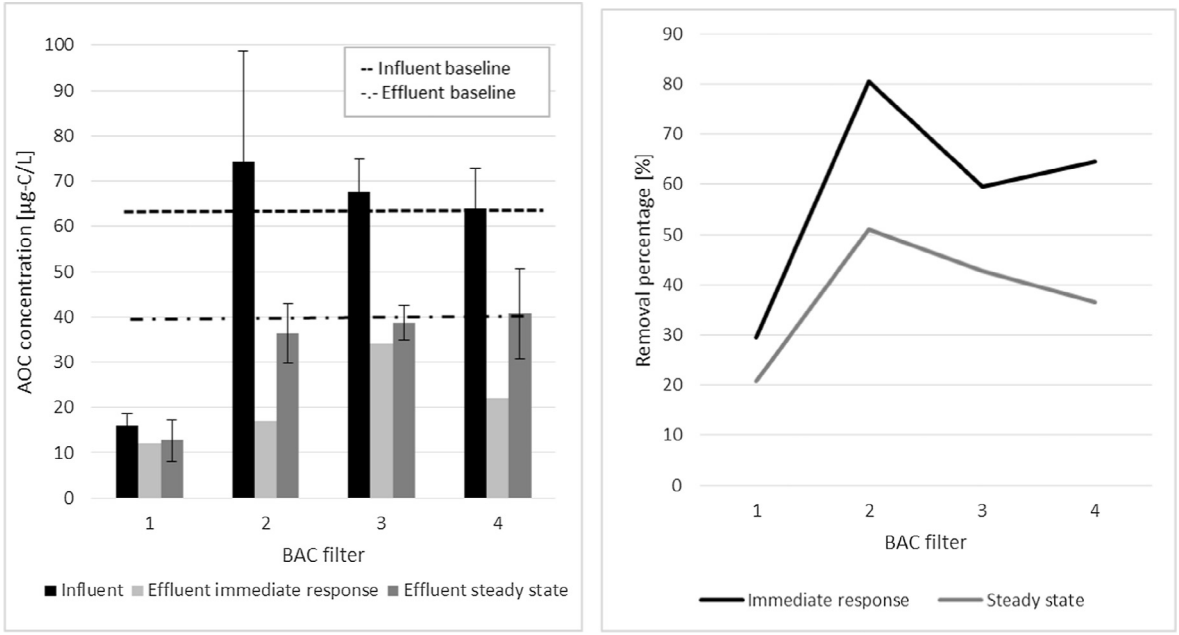

Fig. 5. (a) AOC concentration measured in influent and effluent for both the immediate response and steady state situation (left) and (b) removal percentage AOC per filter for immediate response and steady state situation. The indicated baseline refers to the original situation (BAC 4) and is included for easy comparison.

similar conditions for 5 years. The observed increase might be the result of the sampling period and subsequent seasonal fluctuations. Reactor biomass and ATP on carbon measurements reported by Velten et al. [14] also showed fluctuations occurring in the filter bed after reaching steady state situation.

The highest DOC concentration of all filters was measured in the effluent of BAC 1, which still contained a yellowish color. The water quality of the effluent of BAC 1 showed the importance of a disinfection pre-treatment step before BAC. The addition of phosphate to the influent of BAC 3 resulted on average, in an increased DOC removal. The intact cell concentration in the feed water of both filters was similar, indicating that a higher nutrient concentration resulted in a better performance of the filter, also found by Dhawan et al. [22]. Juhna and Rubulis [18] found similar results for possible phosphate limitation due to pre-treatment with coagulation/flocculation. It would be interesting to investigate if the addition of phosphate to BAC 2 would also have enhanced the DOC removal. In general, the removal percentages found for DOC were similar to the percentages found by Velten et al. [14], even though the influent concentrations in this study were almost 5 times higher.

The effect of intact cell concentration was determined by comparing BAC 2 with BAC 4. The feed water of BAC 2 contained 43% less intact cells, whilst the AOC concentration, representing the bacterial growth potential of the water, was only 9% higher than BAC 4. Contrary to what Liao et al. [13] found, the intact cell concentration in the influent had no determining factor in the performance of the BAC filters.

4.2. Phosphate dosing requirements

The phosphate uptake showed a gradual decrease until a steady state was reached (Fig. 3). The high uptake in the beginning suggested the adsorption of phosphate onto the activated carbon, as seen by several researchers [35,36]. On the other hand, Momba and Cloete [37] described that the uptake of phosphate in a wastewater treatment plant and subsequent removal changes depending on growth state the biomass was in. High phosphate uptake was found during the lag and logarithmic growth phase and low phosphate uptake was found during the stationary phase. Being able to understand the processes taking place might help to understand and develop an optimized dosing scheme.

4.3. Immediate response to change in feed water quality

BAC 2 and 3 showed a different behavior in intact cells directly after the start of the experiment and once steady state was reached. Both filters showed an increase in intact cells in the effluent, whilst this either reduced (BAC 2) or changed to a decrease in intact cells for the steady state situation. In BAC 3 the nutrient balance in the feed water changed by the addition of phosphate. In BAC 2 the significant increase was likely caused by an increase in nutrient concentration (readily available carbon) since the filter was directly fed with ozonated water without any intermediate biological treatment step, in this case pellet softening. The increased biodegradability of the organic matter in the water resulted in an immediate response by the biomass, converting more DOC for both filters. Interestingly the DOC concentration in the effluent of BAC 1 also improved compared to BAC 4. Even though the DOC concentration was the highest in BAC 1 feed water, the immediate response, after start-up of the experiment, showed an increased removal of DOC resulting in the lowest effluent concentration (Fig. 4a). The application of ozone, prior to start experiment, had resulted in higher concentration of DOC with polar groups within the organic molecules, having reduced the adsorption capacity of DOC [38,39]. Thus, even though the BAC filters were in operation for five years, switching the feed water quality from ozonated to non-ozonated water probably resulted in increased (non-polar) DOC adsorption.

The reaction of the biomass and improved adsorption resulted in an improved effluent quality immediately after the water quality of the feed water changed.

5. Conclusions

The main objective of this study was to determine the immediate response of the BAC filters to a rapid change in feed water quality. First, it was shown that with the studied setup it was possible to compare the effects of different pre-treatment steps and subsequent different water qualities on the performance of the BAC filters on the long term adaptation. However, especially the immediate response of the BAC filters on water quality changes was not studied in detail before. It was

observed that all filters were able to mitigate a sudden change in feed water quality, either through improved adsorption or increased activity of the biomass on the filter media. As a result of this resilience against sudden changes, it could therefore be concluded that there is no direct need for very stringent on-line monitoring and continuous adjustments of the feed water quality of the BAC filters.

The long term adaptation of the BAC filters confirmed the need for sufficient nutrients (readily available carbon and phosphate) in the feed water for optimal performance. The influence of intact cells was shown to be limited. Interestingly the storage of phosphate in the BAC filters as a result of adsorption or uptake by the biomass does not necessitate continuous dosing of phosphoric acid. It is recommended to further investigate the optimized dosing scheme.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.seppur.2018.11.072.

References

[1] J.P.v.d. Hoek, J.A.M.H. Hofman, A. Graveland, The use of biological activated carbon filtration for the removal of natural organic matter and organic micropollutants from water, Water Sci. Technol. 40 (9) (1999) 257–264.
[2] L.T.J. van der Aa, A. Magic-Knezev, L.C. Rietveld, J.C. van Dijk, Biomass development in biological activated carbon filters, Mulheim, Germany, 2006a.
[3] W.J. Weber Jr., Preloading of GAC by natural organic matter in potable water treatment systems: Mechanisms, effects and design considerations, J. Water Supply Res. Technol. AQUA 53 (7) (2004) 469–482.
[4] A. Graveland, Application of biological activated carbon filtration at Amsterdam water supply, Water Supply 14 (2) (1996) 233–241.
[5] W. Nishijima, M. Tojo, M. Okada, A. Murakami, Biodegradation of organic substances by biological activated carbon - Simulation of bacterial activity on granular activated carbon, Water Sci. Technol. 26 (9–11) (1992) 2031–2034.
[6] I.C. Escobar, A.A. Randall, J.S. Taylor, Bacterial growth in distribution systems: Effect of assimilable organic carbon and biodegradable dissolved organic carbon, Environ. Sci. Technol. 35 (17) (2001) 3442–3447.
[7] L.T.J. van der Aa, L.C. Rietveld, J.C. van Dijk, Effects of ozonation and temperature on the biodegradation of natural organic matter in biological granular activated carbon filters, Drinking Water Eng. Sci. 4 (1) (2011) 25–35.
[8] P. Servais, G. Billen, P. Bouillot, Biological colonization of granular activated carbon filters in drinking- water treatment, J. Environ. Eng. 120 (4) (1994) 888–899.
[9] V. Lazarova, J. Manem, Biofilm characterization and activity analysis in water and wastewater treatment, Water Res. 29 (10) (1995) 2227–2245.
[10] F. Persson, G. Heinicke, W. Uhl, T. Hedberg, M. Hermansson, Performance of direct biofiltration of surface water for reduction of biodegradable organic matter and biofilm formation potential, Environ. Technol. 27 (9) (2006) 1037–1045.
[11] W. Uhl, R. Gimbel, Dynamic modeling of ammonia removal at low temperatures in drinking water rapid filters, Water Sci. Technol. (2000) 199–206.
[12] D.R. Simpson, Biofilm processes in biologically active carbon water purification, Water Res. 42 (12) (2008) 2839–2848.
[13] X. Liao, R. Zou, C. Chen, B. Yuan, Z. Zhou, H. Ma, X. Zhang, Biomass development in GAC columns receiving influents with different levels of nutrients, Water Sci. Technol.: Water Supply (2016) 1024–1032.
[14] S. Velten, M. Boller, O. Köster, J. Helbing, H.U. Weilenmann, F. Hammes, Development of biomass in a drinking water granular active carbon (GAC) filter, Water Res. 45 (19) (2011) 6347–6354.
[15] F.A. Hammes, T. Egli, New method for assimilable organic carbon determination using flow-cytometric enumeration and a natural microbial consortium as inoculum, Environ. Sci. Technol. 39 (9) (2005) 3289–3294.
[16] F. Hammes, M. Berney, Y. Wang, M. Vital, O. Köster, T. Egli, Flow-cytometric total bacterial cell counts as a descriptive microbiological parameter for drinking water treatment processes, Water Res. 42 (1–2) (2008) 269–277.
[17] D. van der Kooij, A. Visser, W.A.M. Hijnen, Determining the concentration of easily assimilable organic carbon in drinking water, J. Am. Water Works Assoc. 74 (10) (1982) 540–545.
[18] T. Juhna, J. Rubulis, Problem of DOC removal during biological treatment of surface water with a high amount of humic substances, Water Sci. Technol. Water Supply 4 (4) (2004) 183–187.
[19] M.J. Lehtola, I.T. Miettinen, T. Vartiainen, P.J. Martikainen, Changes in content of microbially available phosphorus, assimilable organic carbon and microbial growth potential during drinking water treatment processes, Water Res. 36 (15) (2002) 3681–3690.
[20] A. Sathasivan, S. Ohgaki, K. Yamamoto, N. Kamiko, Role of inorganic phosphorus in controlling regrowth in water distribution system, Water Sci. Technol. (1997) 37–44.
[21] L.T.J. van der Aa, L.J. Kors, A.P.M. Wind, J.A.M.H. Hofman, L.C. Rietveld, Nitrification in rapid sand filter: Phosphate limitation at low temperatures, in: Conference Proceedings Modelling (2006), Delft, the Netherlands, 2002, pp. 37–46.
[22] S. Dhawan, O.D. Basu, B. Banihashemi, Influence of nutrient supplementation on DOC removal in drinking water biofilters, Water Sci. Technol. Water Supply 17 (2) (2017) 422–432.
[23] A.D. Eaton, L.S. Clesceri, E.W. Rice, A.E. Greenberg, M.H. Franson (Eds.), Standard Methods for the Examination of Water and Wastewater, American Public Health Association, Washington D.C., 2005.
[24] D. van der Kooij, W.A.M. Hijnen, Substrate utilization by an oxalate-consuming Spirillum species in relation to its growth in ozonated water, Appl. Environ. Microbiol. 47 (3) (1984) 551–559.
[25] M. Vital, M. Dignum, A. Magic-Knezev, P. Ross, L. Rietveld, F. Hammes, Flow cytometry and adenosine tri-phosphate analysis: Alternative possibilities to evaluate major bacteriological changes in drinking water treatment and distribution systems, Water Res. 46 (15) (2012) 4665–4676.
[26] F. Hammes, F. Goldschmidt, M. Vital, Y. Wang, T. Egli, Measurement and interpretation of microbial adenosine tri-phosphate (ATP) in aquatic environments, Water Res. 44 (13) (2010) 3915–3923.
[27] A. Magic-Knezev, D. van der Kooij, Optimisation and significance of ATP analysis for measuring active biomass in granular activated carbon filters used in water treatment, Water Res. 38 (18) (2004) 3971–3979.
[28] E. Siebel, Y. Wang, T. Egli, F. Hammes, Correlations between total cell concentration, total adenosine tri-phosphate concentration and heterotrophic plate counts during microbial monitoring of drinking water, Drinking Water Eng. Sci. 1 (1) (2008) 1–6.
[29] Matlab, The language of technical computing, version 2009b, The Mathworks Inc., 2009.
[30] A.W.C. van der Helm, L.C. Rietveld, E.T. Baars, P.W.M.H. Smeets, J.C. van Dijk, Modeling disinfection and by-product formation during the initial and the second phase of natural water ozonation in a pilot-scale plug flow reactor, J. Water Supply Res. Technol. AQUA 57 (6) (2008) 435–449.
[31] F. Hammes, N. Boon, M. Vital, P. Ross, A. Magic-Knezev, M. Dignum, Bacterial colonization of pellet softening reactors used during drinking water treatment, Appl. Environ. Microbiol. 77 (3) (2011) 1041–1048.
[32] D. Urfer, P.M. Huck, Measurement of biomass activity in drinking water biofilters using a respirometric method, Water Res. 35 (6) (2001) 1469–1477.
[33] K. Lautenschlager, C. Hwang, F. Ling, W.-T. Liu, N. Boon, O. Köster, T. Egli, F. Hammes, Abundance and composition of indigenous bacterial communities in a multi-step biofiltration-based drinking water treatment plant, Water Res. 62 (2014) 40–52.
[34] P.S. Ross, F. Hammes, M. Dignum, A. Magic-Knezev, B. Hambsch, L.C. Rietveld, A comparative study of three different assimilable organic carbon (AOC) methods: Results of a round-robin test, Water Sci. Technol.: Water Supply (2013) 1024–1033.
[35] P. Kumar, S. Sudha, S. Chand, V.C. Srivastava, Phosphate removal from aqueous solution using coir-pith activated carbon, Sep. Sci. Technol. 45 (10) (2010) 1463–1470.
[36] W. Nishijima, E. Shoto, M. Okada, Improvement of biodegradation of organic substance by addition of phosphorus in biological activated carbon, Water Sci. Technol. 36 (12) (1997) 251–257.
[37] M. Momba, T.E. Cloete, Biomass relationship to growth and phosphate uptake of Pseudomonas fluorescens, Escherichia coli and Acinetobacter radioresistens in mixed liquor medium, J. Ind. Microbiol. 16 (6) (1996) 364–369.
[38] H. Sontheimer, J.C. Crittenden, R.S. Summers, Activated Carbon for Water Treatment, AWWA - DVGW Forschungsstelle Engler Bunte Institut, Karlsruhe, Germany, 1988.
[39] L.T.J. van der Aa, L.C. Rietveld, W.G. Siegers, J.C. van Dijk, Adsorption of natural organic matter and atrazin on granular activated carbon filters, Delft, The Netherlands, 2006b.