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Effects of ozonation and temperature on biodegradation of natural organic matter in biological granular activated carbon filters

L. T. J. van der Aa^{1,2}, L. C. Rietveld², and J. C. van Dijk²

¹Waternet, P.O. Box 94370, 1090 GJ Amsterdam, The Netherlands

²Delft University of Technology, P.O. Box 5048, 2600 GA Delft, The Netherlands

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Correspondence to: L. T. J. van der Aa (rene.van.der.aa@waternet.nl)

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Abstract

Four pilot (biological) granular activated carbon ((B)GAC) filters were operated to quantify the effects of ozonation and water temperature on the biodegradation of natural organic matter (NOM) in (B)GAC filters. Removal of dissolved organic carbon (DOC), assimilable organic carbon (AOC) and oxygen and the production of carbon dioxide were taken as indicators for NOM biodegradation. Ozonation stimulated DOC and AOC removal in the BGAC filters, but had no significant effect on oxygen removal and carbon dioxide production. The temperature had no significant effect on DOC and AOC removal, while oxygen removal and carbon dioxide production increased with increasing temperature. Multivariate linear regression was used to quantify these relations. In summer the ratio between oxygen consumption and DOC removal exceeded the theoretical maximum of $2.5 \text{ g O}_2 \cdot \text{g C}^{-1}$ and the ratio between carbon dioxide production and DOC removal exceeded the theoretical maximum of $3.7 \text{ g CO}_2 \cdot \text{g C}^{-1}$. Bioregeneration of large NOM molecules could explain this excesses and the non-correlation between DOC and AOC removal and oxygen removal and carbon dioxide production. However bioregeneration of large NOM molecules was considered not likely to happen, due to sequestration.

1 Introduction

In the Netherlands the main reasons to use activated carbon (AC) in drinking water treatment are the removal of organic micro pollutants and the removal of substances causing color, taste and odor. Granular activated carbon (GAC) filtration is often preceded by a pre-oxidation step, for example ozonation. Pre-oxidation is used for disinfection, oxidation of micro pollutants and the improvement of color, taste and odor (Kruithof and Maschelein, 1999). An other important effect of pre-oxidation is that it increases the biodegradability of natural organic matter (NOM) (Carlson and Amy, 1987; Hammes et al., 2006; Rietveld et al., 2008; van der Kooij et al., 1989; Yavich et al.,

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2004). This enhances growth of biomass in subsequent treatment steps and results in biological granular activated carbon (BGAC) filtration, also known as biological activated carbon filtration (Graveland, 1994; Jekel, 1979; Sontheimer et al., 1988). In BGAC filters adsorption and biodegradation of NOM occur simultaneously. Adsorption of NOM seriously hinders the adsorption of organic micro pollutants due to pore blocking, competition and pre-loading (Carter et al., 1992; Carter and Weber, 1994; Knappe et al., 1997). When a part of the NOM is biodegraded, adsorption of organic micro pollutants is less hindered. As a result BGAC filtration is found to be more effective than GAC filtration for the removal of NOM (Graveland, 1994; Sontheimer et al., 1988), atrazine (Orlandini, 1999), micro cystines (Wang et al., 2007) and other organic micro pollutants (van der Aa, 2010). In case the biodegradable NOM, produced by the pre-oxidation, is not sufficiently removed in subsequent treatment steps, this will have a negative impact on the bio-stability of the treated water. In the Netherlands drinking water is often distributed without any residual disinfectant. Therefore, much attention is given to biofilm formation and regrowth in the distribution system (Escobar et al., 2001; Servais et al., 2004; van der Kooij, 1992). In BGAC filters biomass has a large influence on the removal of biodegradable NOM and therefore also on the bio-stability.

NOM consists of a mixture of compounds (Leenheer and Croué, 2003). Heterotrophic bacteria take up some of these organic substances. Part of it is used to build up cell material (assimilation) and part of it is oxidized to generate energy for growth and maintenance (dissimilation). During the biodegradation of NOM heterotrophic bacteria consume oxygen and produce carbon dioxide and water (van der Kooij et al., 1982b). The oxygen consumption per amount of biodegraded NOM is constant (Urfer and Huck, 2001). The production of carbon dioxide results in a decrease of pH (de Moel et al., 2006). Each bacterial species has its own preferences for one or more organic compounds. Some of the NOM compounds are biodegraded easily, while others are hardly biodegraded. NOM in natural waters is usually not easily biodegraded, because bacteria already consumed the easily biodegradable part. Pre-oxidation increases the biodegradability of NOM, indicated by increased concen-

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trations of biodegradable dissolved organic carbon (BDOC), easily assimilable organic carbon (AOC) and organic acids, such as oxalate, formiate and acetate (Carlson and Amy, 1987; Hammes et al., 2006; van der Kooij et al., 1989; Yavich et al., 2004). AOC is typically less than 10% of the BDOC in a water sample (Hammes et al., 2006; van der Kooij et al., 1982a; van der Kooij et al., 1989). Biodegradation increases with water temperature and with empty bed contact time. The exhausted (B)GAC filters typically remove 10 to 35% of the NOM from the influent (Graham, 1999; Sontheimer et al., 1988). However, extreme removal ratios were reported from “practically no dissolved organic carbon (DOC) removal” (Juhna and Rubulis, 2004) to 65% (Jekel, 1979). It is assumed that in drinking water usually the carbon source is the limiting factor for biodegradation by heterotrophic bacteria. However, in some cases also phosphate limitation was reported (Juhna and Rubulis, 2004).

The objective of this study was to quantify the effects of ozonation and water temperature on the biodegradation of NOM in (B)GAC filters. One GAC and three BGAC pilot filters were operated treating surface water. After approximately half a year of operation the filters were assumed not to adsorb NOM any more. Removal of DOC, AOC and oxygen and the production of carbon dioxide were taken as indicators for NOM biodegradation. An additional objective was to determine the correlation between these indicators.

2 Materials and methods

2.1 Pilot plant Weesperkarspel: surface water

At WTP Weesperkarspel of Waternet, the water cycle company of Amsterdam and surrounding areas, a pilot plant was operated treating surface water. The raw water was seepage water from the Bethune polder with a high concentration of humic substances (Baghoth et al., 2008; Grefte et al., 2007). The DOC concentration varied between 8 and 10 g C·m⁻³ and the water temperature between 3 and 21 °C. The wa-

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ter was pre-treated in the full-scale plant with coagulation, sedimentation, a reservoir and rapid sand filtration. The pilot plant consisted of 3 ozonation systems, followed by 4 identical gravity filters operated in down flow mode. The influent waters of filters W-GAC8, W-BGAC5, W-BGAC6 and W-BGAC7 received net ozone doses of 0, 0.5, 1.5 and 2.5 g O₃·m⁻³, respectively. The filter media in the (B)GAC filters was Norit GAC830, with a density of 450·10³ g AC·m⁻³. The flow rates were kept constant at 0.88·10⁻³ m·s⁻¹ and the empty bed contact time (EBCT) was 40 min. Figure 1 and Table 1 show an overview of the pilot plant and its characteristics. During operation the (B)GAC filters were backwashed with air and water every 4 to 20 days, depending on filter bed resistance. The columns were monitored on influent, effluent and over the bed height, twice per month. Measured parameters included DOC, UV-extinction at 254 nm, AOC, pH, oxygen and temperature.

2.2 Analytical methods

Ozone was analyzed according to the DPD method (Gilbert, 1981). AOC was measured in duplicate, applying the simultaneous incubation of strains P17 and NOX (van der Kooij et al., 1982b). Other water quality parameters were determined according to standard procedures (Standard Methods, 1998). Carbon dioxide was not analyzed, but calculated from the carbonic equilibrium using pH, conductivity, temperature and the bicarbonate concentration (van Schagen et al., 2008).

2.3 Statistical methods

The Matlab[®] procedure “anova1” (Mathworks, 2007) was used to study differences between datasets. The datasets were considered significantly different from each other when the p-value was smaller than 0.05, which means that the chance that the different datasets do not originate from the same distribution function is more than 95%.

The Matlab[®] procedure “robustfit” (Mathworks, 2007) was used to make linear regressions between different datasets. The procedure “robust” was used to address

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lower weights to outliers. The input variables were considered to correlate significantly with the target variables when the p-value was smaller than 0.05.

Multivariate linear regressions (MLRs) were used to determine which input variable(s) from a group of input variables influenced the target variable. The Matlab[®] procedure “robust” (Mathworks, 2007) was used to address lower weights to outliers. For each target variable MLRs were made for all possible combinations of the input variables. MLRs that included 1 or more input variables with no significant correlation to the target variable (p-value larger than 0.05) were rejected. The MLRs with the lowest root mean sigma error (RMSE) were considered the best MLRs.

3 Results and discussion

Figure 2 shows the concentrations of DOC, AOC and oxygen and the pH of the (B)GAC filters at pilot plant Weesperkarspel at different ozone doses. The minimum, maximum and average influent concentrations are included in Table 1.

3.1 Effects of ozonation and water temperature on water quality

For the water after ozonation MLRs were made for the following target parameters: DOC, AOC, oxygen, carbon dioxide and pH, see Table 2. The following input parameters were used: ozone dose, water temperature, DOC, AOC, oxygen, carbon dioxide and pH, all from the raw water. All target parameters after ozonation correlated significantly with their value before ozonation and were almost linear. The corresponding coefficients were all between 0.92 and 1.0, see Table 2. The DOC concentration after ozonation mainly depended on the DOC concentration in the raw water and decreased slightly with increasing ozone dose. At the highest ozone dose of $2.5 \text{ g O}_3 \cdot \text{m}^{-3}$, resulting in an average ozone/DOC ratio of $0.43 \text{ g O}_3 \cdot \text{g C}^{-1}$, the average DOC removal was $0.17 \text{ g C} \cdot \text{m}^{-3}$ (3%), see Table 1. Other studies reported NOM removal of 3 to 16%, depending on the ozone dose (Sontheimer et al., 1988). Apparently this DOC was

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completely oxidized to carbon dioxide. However, the carbon dioxide concentration did not increase with increasing ozone dose. On the contrary, it decreased with increasing ozone dose, although the correlation was weak. The ozone was dosed via a bubble column, using an ozone-air mixture. It was assumed that the produced carbon dioxide was stripped in the bubble column. No data of carbon dioxide concentrations in the off-gas was available to confirm this hypothesis. The oxygen concentration increased with the ozone dose. Apparently, additional oxygen dissolved from the gas mixture that was dosed in the bubble columns. Also a part of the ozone did not react with substances in the water and was converted into oxygen. The AOC concentration increased with the ozone dose. The increase in AOC of $35 \cdot 10^{-3} \text{ g C} \cdot \text{g O}_3^{-1}$ was within the broad range of values between $20 \cdot 10^{-3}$ and $250 \cdot 10^{-3} \text{ g C} \cdot \text{g O}_3^{-1}$ found by other researchers (Hammes et al., 2006; van der Helm et al., 2007; van der Kooij et al., 1989). There was no significant correlation between AOC concentration after ozonation and the DOC concentrations in the raw water. Other studies showed that the formation of AOC and BDOC during ozonation does increase with the concentration of NOM in the raw water (Carlson and Amy, 1987; van der Helm, 2007). The limited variation in the DOC concentration in the raw water in this study could be a reason for not finding a significant correlation. The ozone decay rate increases with temperature. Therefore, at a constant ozone dose the concentration multiplied by the contact time (CT) for ozone decreases with increasing temperature. Neither the DOC nor the AOC concentration after ozonation depended on the water temperature. Apparently both concentrations depended more on the total amount of ozone dosed than on the CT-value. Van der Helm et al. (2007) also found that the reaction of ozone with NOM was linear with the ozone dose, and not with the CT-value. On the opposite Hammes et al. (2006) found that the formation of AOC did increase with CT.

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3.2 Effects of ozonation and water temperature on biodegradation of NOM in (B)GAC filters

Figure 2 shows that in all (B)GAC filters initial NOM removal was high; 92 to 96% of the DOC and 90 to 100% of the AOC was removed. After 40 days of operation both DOC and AOC concentrations in the effluents increased rapidly according to a typical S-shaped adsorption breakthrough curve. Complete breakthrough was not reached. Instead, after approximately half a year of operation DOC and AOC removal in all (B)GAC filters reached more or less a steady state. It was assumed that the main NOM removal process in this “steady state” situation was biodegradation. To take seasonal effects into account, the biological removal of DOC, AOC, and oxygen and the production of carbon dioxide in the (B)GAC filters were analyzed for a period of 1 year, in the steady state situation, from day 194 to day 559, see Table 3. In this selected period the average DOC removal increased significantly, from 1.2 to 1.8 g C·m⁻³ (20 to 30%), with increasing ozone dose at almost identical DOC influent concentrations. Also the average AOC removal increased significantly with increasing ozone dose. Note that the AOC influent concentrations were increased due to ozonation. However, in none of the filters receiving ozonated water AOC concentrations were reduced to the concentrations before ozonation, see Table 1. The BGAC filters removed up to 70% of the produced AOC. Therefore, ozonation followed by BGAC filtration was expected to have a negative effect on bio-stability. Neither average oxygen consumption nor average carbon dioxide production depended on ozone dose. The significant correlations between the biological removal of DOC, AOC, and oxygen and the production of carbon dioxide were: $\Delta\text{AOC} = 64 \cdot 10^{-3} \cdot \Delta\text{DOC} + 60 \cdot 10^{-3}$ ($R^2 = 0.36$) and $\Delta\text{CO}_2 = -0.79 \cdot \Delta\text{O}_2 + 0.19$ ($R^2 = 0.13$), see Fig. 3.

MLRs were made for the biological removal of DOC, AOC, and oxygen and the production of carbon dioxide in the (B)GAC filters, again for the selected period of one year. Different sets of input variables were used: a) water quality after ozonation (ozone dose was not used), b) water quality after ozonation + ozone dose, c) raw water quality +

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ozone dose, and d) water temperature + ozone dose. When only the water quality variables after ozonation were used as input parameters (set a), 3 MLRs showed a negative R^2 . This indicates that this set was severely influenced by outliers. Therefore, this set was excluded for further examination. Adding the ozone dose as an input variable (set b) reduced this problem. The ozone dose provides valuable information to predict biodegradation in (B)GAC filters. Using the water quality variables from the raw water instead of those from the water after ozonation (set c) increased the RMSEs, meaning that the quality of the MLRs decreased. This is the result of additional noise in the input variables caused by the ozonation. The simplest set (d) only used the temperature and the ozone dose as input. Figure 4 visualizes the results of the MLRs for set d. The results of the MLRs for sets b, c and d are presented in Table 4.

Figure 3, Table 4 and Fig. 4 demonstrate that the removal of DOC and AOC correlated with each other and increased with increasing ozone dose. They were not significantly influenced by the water temperature. Oxygen consumption and carbon dioxide production correlated with each other and increased with increasing temperature. They were not significantly influenced by the ozone dose. The removal of DOC and AOC did not correlate significantly with the removal of oxygen and the production of carbon dioxide.

For complete oxidation of “average NOM” with a C:H:O ratio of 1:1.1:0.7 (Uhl, 2000) approximately $2.5 \text{ g O}_2 \cdot \text{g C}^{-1}$ is needed. The molecular mass of carbon dioxide is $44 \text{ g} \cdot \text{mol CO}_2^{-1}$ and the molecular mass of carbon is $12 \text{ g} \cdot \text{mol C}^{-1}$. When all NOM carbon would be converted into carbon dioxide, this would produce $44/12=3.7 \text{ g CO}_2 \cdot \text{g C}^{-1}$. Figure 5 shows that both the ratio between oxygen and DOC removal and the ratio between carbon dioxide production and DOC removal increased with increasing temperatures. In summer more oxygen was consumed and more carbon dioxide was produced than needed for complete NOM oxidation. This was also observed for other BGAC filters by other researchers (Jekel, 1979; Sontheimer et al., 1988).

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When biodegradation of NOM from the water phase is assumed to be the only relevant process for run times longer than 194 days, the removal DOC, AOC and oxygen and the production of carbon dioxide should: a) correlate with each other, b) increase with increasing temperature, c) increase with increasing ozone dose, and d) the amount of oxygen removed should not exceed $2.5 \text{ g O}_2 \cdot \text{g C}^{-1}$ and the amount of carbon dioxide produced should not exceed $3.7 \text{ g CO}_2 \cdot \text{g C}^{-1}$. This was not the case. It was suggested that, besides biodegradation of NOM from the water phase, the following mechanisms were important: decrease of NOM reduction degree by ozonation, degassing of (B)GAC filters, biomass production and lysis, temperature effects on NOM adsorption and bioregeneration.

3.2.1 Decrease of NOM reduction degree by ozonation

The average ratio between oxygen removal and DOC removal decreased from 3.2 to $1.9 \text{ g O}_2 \cdot \text{g C}^{-1}$ with increasing ozone dose, see Table 3. However, the significance was relatively low (p-value 0.06). Due to the reaction of NOM and ozone more oxygen atoms are incorporated in the NOM molecules, resulting in a decrease of the reduction degree of the NOM. For biodegradation of NOM with a lower reduction degree less oxygen is consumed (Uhl, 2000). From his data Uhl derived that due to this oxidation the reduction degree of NOM reduced by 0.26 to 0.48 in four different (B)GAC filters, corresponding with an oxygen uptake of 0.13 to $0.24 \text{ g O}_2 \cdot \text{g C}^{-1}$. This is less than the observed difference of $3.2 - 1.9 = 1.3 \text{ g O}_2 \cdot \text{g C}^{-1}$ found in this study.

3.2.2 Degassing of (B)GAC filters

The average ratio between carbon dioxide production and DOC removal was between -2.7 and -0.4 for all (B)GAC filters and did not depend on ozone dose, see Table 3. As 3.7 is the ratio between the molecular mass of carbon dioxide ($44 \text{ g} \cdot \text{mol CO}_2^{-1}$) and carbon ($12 \text{ g} \cdot \text{mol C}^{-1}$), this means that not all removed NOM carbon was found as carbon dioxide in the effluent. For example in filter W-GAC8 only $2.4 \cdot 12 / 44 = 65\%$ of the bio-

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logically removed NOM carbon was found as carbon dioxide in the effluent. For filters W-BGAC5, W-BGAC6 and W-BGAC7 this was 11, 38 and 74%, respectively. Ozone was dosed at a water depth of 5 m, while the supernatant water in the (B)GAC filters was at atmospheric pressure. The supernatant water was supersaturated with gas.

5 In summer frequently gas bubbles were observed in the (B)GAC filters. The escaping gas contained relatively high amounts of carbon dioxide (2%). No information on the amounts of gas escaping was available to assess the effect on the carbon balance. However, the gas was escaping at atmospheric pressure, so the density was approximately $1.2 \cdot 10^3 \text{ g} \cdot \text{m}^{-3}$. The concentration of carbon dioxide carbon was approximately $1.2 \cdot 10^{-3} \cdot 2\% \cdot 12/44 = 6.5 \text{ g C} \cdot \text{m}^{-3}$. For example for filter W-GAC8 it was demonstrated that 35 of the NOM carbon was not found as carbon dioxide in the effluent. If this was caused by degassing of the filters $0.35 \cdot 1.2 \cdot 0.42/6.5 = 0.065 \text{ m}^3$ of gas per m^3 of treated water should have escaped from the filters. At a surface loading of $8.7 \cdot 10^{-4} \text{ m} \cdot \text{s}^{-1}$ this corresponded with 4.9 m^3 gas per m^2 filter bed per day, which is not realistic.

15 3.2.3 Biomass production and lysis

A part of the carbon is converted into biomass. This biomass can either accumulate in the filter bed or leave the filter with the effluent or with the backwash water. Also it is possible that the biomass dies, disintegrates and serves as a carbon source for other micro organisms (lysis, predation). During additional filter runs the maximum observed change rate in biomass activity, averaged over the filter bed, was $4 \cdot 10^{-9} \text{ g adenosine triphosphate (ATP)} \cdot \text{g AC}^{-1} \cdot \text{day}^{-1}$ measured according to the method described by Magic-Knezev and van der Kooij (2004) (data not shown). At an EBCT of 40 minutes, an AC density of $450 \cdot 10^3 \text{ g AC} \cdot \text{m}^{-3}$ and a biomass carbon/ATP ratio of 250 (Magic-Knezev and van der Kooij, 2004) this rate corresponds with a change in DOC concentration in the water phase of $4 \cdot 10^{-9} \cdot 250 \cdot 450 \cdot 10^3 / ((24 \cdot 3600) / (40 \cdot 60)) = 0.013 \text{ g C} \cdot \text{m}^{-3}$, which is only about 1% of the DOC removal in the steady state. Therefore, the production and lysis of biomass were not sufficient to explain the discrepancy between NOM removal and carbon dioxide production.

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3.2.4 Temperature effects on NOM adsorption

From a theoretical point of view, adsorption should decrease with increasing temperature (Schreiber et al., 2007). This was experimentally shown for some individual compounds, e.g. phenol and *o*-cresol (Abuzaid and Nakhla, 1997). However, experiments showed increasing NOM adsorption with increasing water temperature (Schreiber et al., 2005; Schreiber et al., 2007; Summers and Roberts, 1988). For Weesperkarspel raw water the adsorption isotherms were only determined at 12 °C (data not shown). Therefore, the effect of temperature on the adsorption isotherm could not be assessed. It is possible that at higher temperatures adsorption decreased, or even completely stopped, and simultaneously biodegradation of NOM increased. The oxygen consumption and carbon dioxide production would increase in summer, while the DOC and AOC removal would remain fairly constant. The hypothesis that NOM adsorbs in winter and is biodegraded in summer can not explain why in summer the oxygen consumption exceeded 2.5 g O₂·g C⁻¹ and the carbon dioxide production exceeded 3.7 g CO₂·g C⁻¹. This hypothesis could only partly explain the discrepancy between DOC and AOC removal on one side and oxygen removal and carbon dioxide production on the other side.

3.2.5 Bioregeneration

Bioregeneration of AC is biodegradation of adsorbed NOM, which results in a decrease of the NOM loading on the AC (Sontheimer et al., 1988). During biodegradation of this NOM oxygen is consumed and carbon dioxide is produced. The NOM that is biodegraded originates from the AC, and not from the water phase. An increase of oxygen consumption and carbon dioxide production is possible, without any effect on the DOC and AOC removal from the water phase. However, NOM from Weesperkarspel raw water mainly consists of large humic acid molecules (Bagtho et al., 2008; Grefte et al., 2007). Even after an ozone dose of 2.5 g O₃·m⁻³ the average amount of small humic acids, indicated by the AOC concentration, was only 2% of the total NOM, see Table 1.

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Bioregeneration of large humic molecules was not likely to happen, due to sequestration of the molecules (Kraai, 2001). Bioregeneration of small humic acids may be possible, but because of the limited concentrations the effect is negligible. For example: assume that in winter $0.1 \text{ g acetate-C} \cdot \text{m}^{-3}$ adsorbs onto the BAC during a period of 6 months and in summer $0.1 \text{ g acetate-C} \cdot \text{m}^{-3}$ is biodegraded from the water phase, plus all adsorbed AOC from the winter period is biodegraded. When for complete oxidation of 1 g acetate-C of AOC 2.5 g oxygen is needed, the extra amount of oxygen consumed in summer would be $0.2 \cdot 2.5 = 0.5 \text{ g O}_2 \cdot \text{m}^{-3}$, which is only approximately 10% of the difference in oxygen consumption in summer and winter. If bioregeneration of larger NOM molecules would be possible, the amounts of bioregeneration could be the tenfold and could explain the high ratios between oxygen and DOC removal and carbon dioxide production and DOC removal in summer. However, bioregeneration of large NOM molecules is not likely to happen. No evidence for bioregeneration was found.

4 Conclusions

In this study the influence of ozonation on the biodegradation of NOM in (B)GAC filters was assessed in four pilot (B)GAC filters. To study a period with biodegradation as the main NOM removal process and to take seasonal effects into account, the filters were judged for a period of one year, from day 194 to day 559.

The main effect of the ozonation was the increase of the AOC concentration in the (B)GAC influents by $35 \cdot 10^{-3} \text{ g acetate-C} \cdot \text{g O}_3^{-1}$. Ozonation also resulted in limited decreases of DOC and oxygen concentrations and in limited increases of pH and (calculated) carbon dioxide concentration.

The BGAC filters receiving ozonated water removed up to 70% of the produced AOC. However, in none of the BGAC filters AOC concentrations were reduced to the concentrations before ozonation. Therefore, pre-oxidation followed by BGAC filtration was expected to have a negative effect on bio-stability. Ozonation also stimulated the removal

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of DOC in the (B)GAC filters. At the highest ozone dose of $2.5 \text{ g O}_3 \cdot \text{m}^{-3}$, resulting in an average ozone/DOC ratio of $0.43 \text{ g O}_3 \cdot \text{g C}^{-1}$, the DOC removal was 30% versus 20% in the filter receiving non-ozonated water. The removal of DOC and AOC correlated with each other and increased with increasing ozone dose. They were not significantly influenced by the water temperature. Oxygen consumption and carbon dioxide production correlated with each other and increased with increasing temperature. They were not significantly influenced by the ozone dose. The removal of DOC and AOC did not correlate significantly with the removal of oxygen and the production of carbon dioxide. This non-correlation indicates that besides biodegradation other processes were relevant for NOM removal in the filters. Theoretical calculations demonstrated that the build up of biomass, degassing of (B)GAC filters, the decrease in NOM reduction degree and the temperature effects on NOM adsorption could only partly explain the non-correlation between DOC and AOC removal and oxygen removal and carbon dioxide production. Bioregeneration of large NOM molecules could explain this non-correlation, but was considered not likely to happen, due to sequestration. No evidence for bioregeneration was found.

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Table 1. Characteristics and influent water qualities pilot plant Weesperkarspel.

Column	W-GAC8	W-BGAC5	W-BGAC6	W-BGAC7
Column diameter [m]	0.25	0.25	0.25	0.25
Bed height [m]	2.10	2.10	2.10	2.10
Carbon type [-]	GAC830	GAC830	GAC830	GAC830
Operation time [s]	586-24-3600	600-24-3600	559-24-3600	586-24-3600
Flow [$\text{m}^3 \cdot \text{s}^{-1}$]	$4.3 \cdot 10^{-5}$	$4.3 \cdot 10^{-5}$	$4.3 \cdot 10^{-5}$	$4.3 \cdot 10^{-5}$
Surface load [$\text{m} \cdot \text{s}^{-1}$]	$8.7 \cdot 10^{-4}$	$8.7 \cdot 10^{-4}$	$8.7 \cdot 10^{-4}$	$8.8 \cdot 10^{-4}$
EBCT [s]	40-60	40-60	40-60	40-60
Net ozone dose [$\text{g O}_3 \cdot \text{m}^{-3}$]	–	0.5	1.5	2.5
Temp [$^{\circ}\text{C}$]	12.8 (3.0–20.6)	12.2 (3.6–20.6)	11.6 (3.8–20.6)	11.6 (3.8–20.6)
pH [–]	7.7 (7.3–7.9)	7.7 (7.3–7.8)	7.7 (7.1–7.8)	7.7 (7.6–7.8)
DOC [$\text{g C} \cdot \text{m}^{-3}$]	6.0 (5.0–6.9)	6.0 (5.2–7.6)	5.9 (4.9–7.5)	5.8 (4.9–6.7)
UV254 [m^{-1}]	14.4 (11.7–17.8)	13.5 (11.6–16.9)	10.8 (5.4–14.4)	8.5 (6.4–10.4)
Oxygen [$\text{g O}_2 \cdot \text{m}^{-3}$]	10.0 (7.9–12.9)	11.0 (8.3–14.2)	11.1 (7.4–13.7)	11.6 (9.5–14.3)
Carbon dioxide [$\text{g CO}_2 \cdot \text{m}^{-3}$]	8.1 (4.6–10.7)	7.7 (0.1–15.7)	8.6 (5.8–39.7)	7.2 (5.5–10.1)
AOC [$10^{-3} \text{g Acetate-C} \cdot \text{m}^{-3}$]	11.5 (5.7–32)	35.0 (16–99)	85.0 (40–199)	114.1 (42–265)

Notes water quality parameters: table values are average concentrations; values between parenthesis are minimum and maximum concentrations; carbon dioxide concentrations calculated according (van Schagen et al., 2008).

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Table 2. MLRs water quality parameters after ozonation.

MLR – Robust Input variables: O ₃ , temp, [DOC] _{raw} , [AOC] _{raw} , [O ₂] _{raw} , [CO ₂] _{raw}	<i>R</i> ²	RMSE
[DOC] _{inf} = 0.51 + 0.92·[DOC] _{raw} – 0.070·O ₃	0.60	0.14
[AOC] _{inf} = 1.0·10 ^{–3} + 1.0·[AOC] _{raw} + 35·10 ^{–3} ·O ₃	0.62	10·10 ^{–3}
[O ₂] _{inf} = 0.46 + 0.98·[O ₂] _{raw} + 0.57·O ₃	0.86	0.49
[CO ₂] _{inf} = 0.44 + 0.94·[CO ₂] _{raw} – 0.32·O ₃	0.15	0.35
pH _{inf} = 0.17 + 0.98·pH _{raw} + 0.014·O ₃	0.48	0.013

Notes: _{raw} indicates raw water, i.e. water before ozonation and _{inf} indicates (B)GAC influent, i.e. water after ozonation.

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Table 3. Biological conversions in (B)GAC pilot filters.

Column	W-GAC8	W-BGAC5	W-BGAC6	W-BGAC7
Net ozone dose [$\text{g O}_3 \cdot \text{m}^{-3}$]	0	0.5	1.5	2.5
ΔDOC [$\text{g C} \cdot \text{m}^{-3}$]	−1.2 (−20%)	−1.3 (−21%)	−1.5 (−25%)	−1.8 (−30%)
ΔAOC [$10^{-3} \text{ g acetate} \cdot \text{C} \cdot \text{m}^{-3}$]	+2.7 (+21%)	−12.3 (−35%)	−68.3 (−73%)	−85.4 (−68%)
ΔO_2 [$\text{g O}_2 \cdot \text{m}^{-3}$]	−3.3 (−54%)	−3.4 (−55%)	−3.6 (−59%)	−3.5 (−59%)
ΔCO_2 [$\text{g CO}_2 \cdot \text{m}^{-3}$]	+2.6 (+33%)	+3.4 (+44%)	+0.9 (+10%)	+2.7 (+39%)
$\Delta\text{O}_2 \cdot \Delta\text{DOC}^{-1}$ [$\text{g O}_2 \cdot \text{g C}^{-1}$]	3.2	2.8	2.3	1.9
$\Delta\text{CO}_2 \cdot \Delta\text{DOC}^{-1}$ [$\text{g CO}_2 \cdot \text{g C}^{-1}$]	−2.4	−2.7	−0.4	−1.4

Notes: Table values are average difference in (B)GAC influent and effluent concentrations $C_{\text{eff}} - C_{\text{inf}}$ during day 194 to day 559; percentage values are average relative removal percentages $(C_{\text{eff}} - C_{\text{inf}})/C_{\text{inf}}$.

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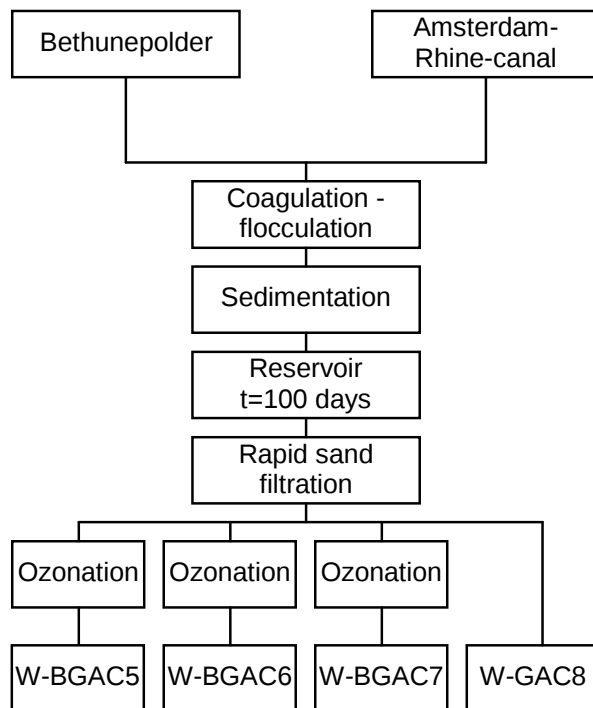
Table 4. MLRs biological conversions in (B)GAC pilot filter.

MLR – Robust – set b Input variables: O_3 , temp, $[DOC]_{inf}$, $[AOC]_{inf}$, $[O_2]_{inf}$, $[CO_2]_{inf}$	R^2	RMSE
$\Delta[DOC] = 1.2 - 0.39 \cdot [DOC]_{inf} - 0.26 \cdot O_3$	0.53	0.22
$\Delta[AOC] = 13 \cdot 10^{-3} - 0.90 \cdot [AOC]_{inf} - 4.0 \cdot 10^{-3} \cdot O_3$	0.96	$6.3 \cdot 10^{-3}$
$\Delta[O_2] = 173 - 0.57 \cdot temp - 0.27 \cdot [O_2]_{inf} - 20 \cdot pH_{inf} - 1.5 \cdot [CO_2]_{inf}$	-7.1	0.50
$\Delta[CO_2] = -5.4 + 0.36 \cdot temp + 0.54 \cdot [O_2]_{inf} - 0.18 \cdot [CO_2]_{inf} - 0.57 \cdot O_3$	0.46	0.94
MLR – Robust – set c Input parameters: O_3 , temp, $[DOC]_{raw}$, $[AOC]_{raw}$, $[O_2]_{raw}$, $[CO_2]_{raw}$		
$\Delta[DOC] = -1.2 - 0.23 \cdot O_3$	0.34	0.25
$\Delta[AOC] = 179 \cdot 10^{-3} - 29 \cdot 10^{-3} \cdot [DOC]_{raw} - 34 \cdot 10^{-3} \cdot O_3$	0.58	$15 \cdot 10^{-3}$
$\Delta[O_2] = 7.3 - 0.38 \cdot temp - 0.48 \cdot [O_2]_{raw} - 0.13 \cdot [CO_2]_{raw}$	0.88	0.53
$\Delta[CO_2] = -0.58 + 0.27 \cdot temp$	0.20	1.0
MLR – Robust – set d Input parameters: O_3 , temp		
$\Delta[DOC] = -1.2 - 0.23 \cdot O_3$	0.34	0.25
$\Delta[AOC] = 0.92 \cdot 10^{-3} - 29 \cdot 10^{-3} \cdot O_3$	0.49	$16 \cdot 10^{-3}$
$\Delta[O_2] = 0.20 - 0.27 \cdot temp$	0.73	0.64
$\Delta[CO_2] = -0.58 + 0.27 \cdot temp$	0.20	1.0

Notes: $_{raw}$ indicates raw water, i.e. water before ozonation; $_{inf}$ indicates (B)GAC influent, i.e. water after ozonation; Δ indicates the difference in (B)GAC influent and effluent concentrations $C_{eff} - C_{inf}$ during day 194 to day 559.

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**Fig. 1.** Flow scheme pilot plant Weesperkarspel.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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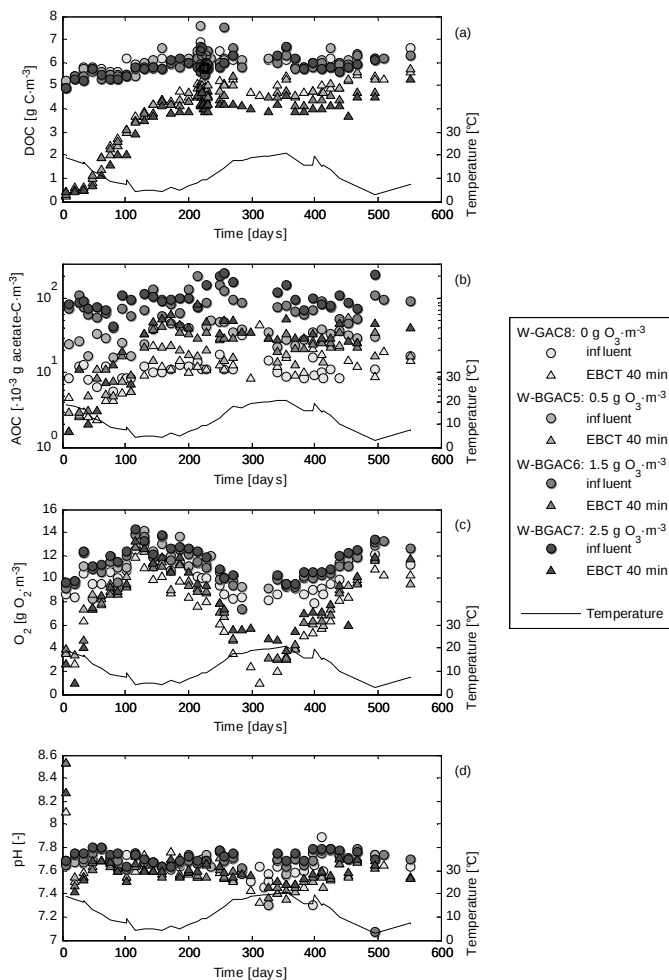


Fig. 2. DOC (a), AOC (b), oxygen (c) and pH (d) in Weesperkarspel pilot (B)GAC filters.

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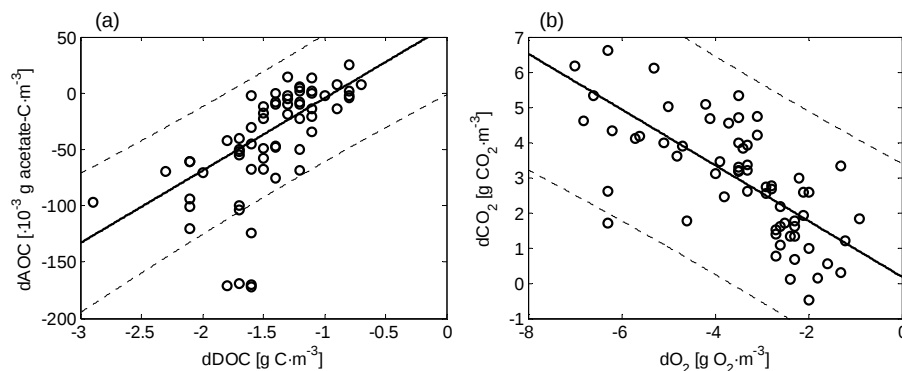


Fig. 3. Linear regressions plus 95% confidence intervals for data from all 4 (B)GAC filters: $\Delta\text{AOC} = 64 \cdot 10^{-3} \cdot \Delta\text{DOC} + 60 \cdot 10^{-3}$ ($R^2 = 0.36$) (a) and $\Delta\text{CO}_2 = -0.79 \cdot \Delta\text{O}_2 + 0.19$ ($R^2 = 0.13$) (b).

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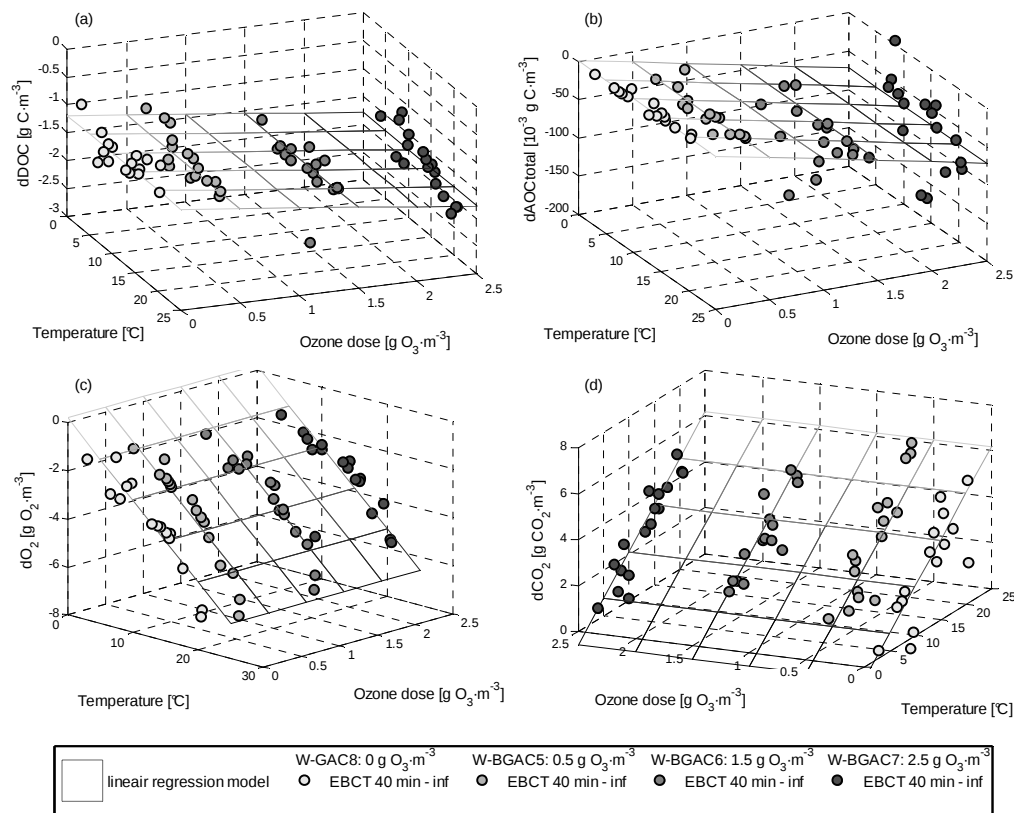


Fig. 4. MLRs for biological conversions of DOC (a), AOC (b), oxygen (c) and carbon dioxide (d) as function of ozone dose and temperature from day 194 to 559 (set d).

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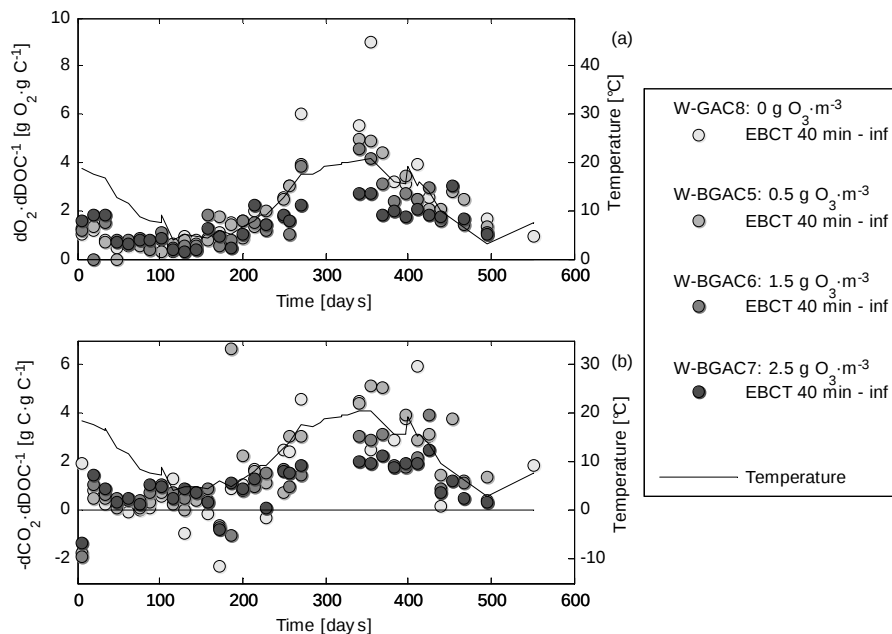


Fig. 5. Oxygen to DOC removal ratio **(a)** and carbon dioxide production to DOC removal ratio **(b)**.

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