Authors' reply to comments anonymous referee 2

The critical comments by the reviewer are appreciated. Thanks to the referees' suggestions additional literature was studied, resulting in new ideas. The conclusion that bioregeneration was 'not likely' in the (biological) granular activated carbon filters, was changed into that it was 'likely' that bioregeneration occurred.

Specific comments anonymous referee 2, including authors' reply in italic.

Obviously biological activity takes place and in the fourth filter and in any case the biological contribution on NOM removal is not well documented. From experimental data presented it is not obvious the effect of ozone on NOM removal and the authors try a lot to reach some conclusions. Conclusions are not clear and cannot be supported efficiently.
 # Various hypothetical interfering processes were discussed. Because in the experiments no distinction could be made between the fates of NOM, only theoretical calculations were made.

The results from additional literature research indicate that bioregeneration of large NOM molecules is possible. Therefore, all sections about bioregeneration were revised:

In the section 'Summary':
"The production and loss of biomass, the degassing of (B)GAC filters, the decrease in the NOM reduction degree and the temperature effects on NOM adsorption could only partly explain these excesses and the non-correlation between DOC and AOC removal and oxygen consumption and carbon dioxide production. It was demonstrated that bioregeneration of NOM could explain the excesses and the non-correlation. Therefore, it was likely that bioregeneration of NOM did occur in the (B)GAC pilot filters."

In the section 'Results and discussion':
"Bioregeneration
Bioregeneration of AC is biodegradation of (previously) adsorbed NOM, which results in a decrease in the NOM loading on the AC (Sontheimer et al., 1988). Several authors described two possible mechanisms for bioregeneration. The first hypothesis is that biomass on the external AC surface takes up substrate. Therefore, the concentration of the substrate on the external AC surface becomes smaller than the internal equilibrium concentration. This causes diffusion of the substrate from the internal pores towards the external AC surface, where it is biodegraded. The concentration inside the pores decreases, which results in desorption. The AC is available for adsorption again: it has been bioregenerated. The second hypothesis is that the biomass releases extracellular enzymes that enter the meso-pores of the AC; micro-pores are believed to be too small for the exo-enzymes to enter. The exo-enzymes convert part of the adsorbed substrate into less adsorbable products. These products desorb and diffuse from the internal pores towards the external AC surface, where they are biodegraded. Again, the AC is bioregenerated. In both hypotheses, both desorption and biodegradation are conditions for bioregeneration (Aktas and Çeçen, 2007; Klimenko et al., 2003; Walker and Weatherley, 1998).

During bioregeneration, oxygen is consumed and carbon dioxide is produced. The NOM that is biodegraded originates from the adsorbed phase. An increase in oxygen consumption and carbon dioxide production is possible, without any effect on the measured DOC and AOC concentrations in the filter effluent. During

complete oxidation of 1 g C 2.6 g oxygen is needed and 3.7 g carbon dioxide is produced. Assume that in winter, during a period of 6 months, 1.0 g C·m^{-3} NOM adsorbed onto the AC and that 0.2 g C·m^{-3} NOM was biodegraded. The oxygen consumption and the carbon dioxide production, per amount of NOM removed from the water phase, would have been 0.2·2.6/1.2 = 0.4 g O_2·g C^{-1} and 0.2·3.7/1.2 = 0.6 g CO_2·g C^{-1}. Assume that in summer, during a period of 6 months, no NOM adsorbed, that 1.2 g C·m^{-3} NOM was biodegraded from the water phase and that all adsorbed NOM from the previous winter period was biodegraded. The oxygen consumption and the carbon dioxide production, per amount of NOM removed from the water phase, would have been (1.2+1)·2.6/1.2 = 4.8 g O_2·g C^{-1} and (1.2+1)·3.7/1.2 = 6.8 g CO_2·g C^{-1}. These figures correspond well to the measured results from the pilot experiment, as seen in Figure 5.

AC bioregeneration was reported for different specific compounds in (industrial) waste waters (Aktas and Çeçen, 2007; Klimenko et al., 2003; Walker and Weatherley, 1998). Although no hard evidence was found, some researchers suggested that bioregeneration of NOM in drinking water is possible (Sontheimer et al., 1988). Both desorption and biodegradation of the compounds are conditional for bioregeneration. It is obvious that a part of the NOM in the pilot (B)GAC filters was biodegradable and adsorbable (Figure 2 and Table 3). For batch experiments, desorption of 4% to 58% of previously adsorbed NOM was reported. The percentage of NOM desorption depended on the type of NOM and on the type of AC (Yapsakli et al., 2009), Therefore, in theory, the conditions for biodegradation and for desorption can be met. Because in summer the reported ratio between oxygen consumption and DOC removal exceeded 2.6 g O_2·g C^{-1} and the ratio between carbon dioxide production and DOC removal exceeded 3.7 g CO_2·g C^{-1}, it was likely that bioregeneration of NOM did occur in the (B)GAC pilot filters."

In the section 'Conclusions':
"Bioregeneration of NOM could explain the excesses and the non-correlation. Therefore, it was likely that bioregeneration of NOM did occur in the (B)GAC pilot filters.

It is recommended that adsorption, desorption and biodegradation experiments be performed with labeled ^{14}C-glucose (Servais et al., 1994), or if possible with larger (both biodegradable and non-biodegradable) ^{14}C-NOM molecules. This will make it possible to determine the fate of NOM and to quantify relevant processes in BGAC filtration. Possibly, hard evidence for bioregeneration of NOM will be found."

A discussion part of the cost of the proposed method, especially for higher ozone concentrations, is missing.
 # It was not the objective of this paper to discuss the costs of BGAC filtration. Raw water quality, pre-treatment (for example ozone) and the desired water quality have an enormous impact on the design of BGAC filtration (investment costs) and on the filter life time (operational costs). Some cost issues regarding the combination of ozone and BGAC filtration at the Weesperkarspel water treatment plant were reported elsewhere (van der Helm et al., 2008).

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