Fate of bulk organic carbon and bromate during indirect water reuse involving ozone and subsequent aquifer recharge

U. Hübner, S. Kuhnt, M. Jekel and J. E. Drewes

ABSTRACT

Long-term column studies with ozonated secondary effluent were conducted to evaluate the removal of dissolved organic carbon (DOC) and to assess formation and stability of the oxidation by-product bromate in combined ozonation and managed aquifer recharge (MAR) systems. DOC removal during simulated MAR was continuously increased from approximately 20 to 40% by pretreatment with ozone indicating the high potential of this combination to remove disinfection by-product (DBP) precursors. This improvement in DOC removal was likely caused by transforming DOC into fractions that are more amenable to biodegradation, as well as the supply of dissolved oxygen favouring aerobic degradation processes. Bromate formation during ozonation of secondary effluent from the Berlin-Ruhleben WWTP exceeded the limit of the EU Drinking Water Directive at a specific ozone consumption of >0.9–1.0 mg O_3/mg DOC_0. During oxic infiltration, no significant removal of bromate could be observed. Under anoxic conditions, bromate concentration was efficiently reduced. Additional biodegradation tests in small-scale columns indicated a simultaneous consumption of nitrate and bromate as electron acceptors.

Key words | bromate, dissolved organic carbon, managed aquifer recharge, ozonation, riverbank filtration

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INTRODUCTION

In recent years, there has been growing concern regarding the occurrence of trace organic compounds (TrOCs) in the aquatic environment (Schwarzenbach *et al.* 2006). Conventional wastewater treatment plants (WWTPs) are not primarily designed for their removal and polar and non-biodegradable TrOCs in particular are not efficiently eliminated and end up in receiving waters with concentrations in the ng/L to µg/L-range (Clara *et al.* 2005; Joss *et al.* 2006; Reemtsma *et al.* 2006), where they can pose potential risks for aquatic organisms and downstream drinking water supplies. Chronic exposure to compounds with estrogenic potency, for example, has been reported to cause feminization of male fish (Tyler *et al.* 2005). Managed aquifer recharge (MAR) systems are engineered processes that use the aquifer's natural capacity for water treatment and storage, such as bank filtration, artificial groundwater recharge and soil aquifer treatment, infiltrating reclaimed water. They are highly effective for the removal of conventional contaminants and can also efficiently retain many TrOCs (Grünheid *et al.* 2005; Hoppe-Jones *et al.* 2010), however, some highly persistent TrOCs can still be detected in riverbank filtrate (Drewes *et al.* 2003) and even reach drinking water (Ternes *et al.* 2002).

Ozonation is a powerful process for the removal of many TrOCs from secondary effluents (Hollender *et al.* 2009) and its combination with MAR was proposed by several researchers as a multiple barrier system for TrOCs (Hübner *et al.* 2012; Yoon *et al.* 2013). In addition, ozonation may enhance biodegradation of bulk organic carbon in MAR systems by supplying excess oxygen and pre-oxidation

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of organic matter (Schumacher *et al.* 2004; Hübner *et al.* 2012). Furthermore, MAR may function as an efficient barrier for potentially toxic transformation products from TrOCs and oxidation by-products such as N-Nitrosodimethylamine (NDMA) or bromate. Microbial degradation of transformation products from ozone reactions has been reported previously in several studies (Scheurer *et al.* 2012; Hübner *et al.* 2014). Also biodegradation of NDMA was shown to be efficient in MAR systems (Drewes *et al.* 2006). To our knowledge, the biological removal of bromate in MAR systems has not been addressed in previous studies. Hübner *et al.* (2012) observed no significant degradation in an oxic slow sand filter. However, results from experiments under anoxic conditions demonstrate that bromate can be used as an electron acceptor for anaerobic growth (Kirisits *et al.* 2001; Van Ginkel *et al.* 2005; Downing & Nerenberg 2007). Cometabolic bromate removal by nitrate reductase, but also bromate specific reduction pathways, were suggested (Davidson *et al.* 2011).

In this study we assessed the effect of ozonation on the removal of bulk organic carbon in long-term column studies, with a special focus on elucidating the effects of pre-oxidation of organic matter and a supply of excess oxygen for subsequent aerobic biodegradation processes. In addition, the removal of bromate was investigated in different column systems under both oxic and anoxic conditions.

EXPERIMENTS

Description of long-term column studies

Four sand columns (SC1 SC4, length 100 cm, inner diameter 14 cm) were operated at ambient temperature (22 °C) under saturated flow conditions against gravity. Columns were filled with technical sand (particle size: 0.7–1.2 mm) and adapted to the respective feed water for approximately 2 months prior to the first sampling campaign. Secondary effluent from the Berlin-Ruhleben WWTP (dissolved organic carbon (DOC): 11.5 ± 1.3 mg/L) was collected weekly as feed water for the experiments. Bromide was not measured in this study, but previous analyses revealed an average concentration of 112 µg/L. Columns SC1 and SC2 were continuously operated as reference without additional pretreatment and with ozonated WWTP effluent (using a specific ozone consumption of 0.74 ± 0.11 mg O_3/mg DOC_0), respectively. Preozonation for SC3 was conducted with different specific ozone consumption (ranging from 0.7 to 1.0 mg O_3/mg DOC_0). Subsequently, the feed water for column SC3 was continuously purged with nitrogen in a bubble column to evaluate subsequent bromate removal under anoxic conditions. The addition of hydrogen peroxide was tested in column SC4 to establish oxic conditions throughout the column. In addition, a spiking experiment with 100 µg/L bromate was conducted using columns SC2 and SC3 over a period of 4 months (September–December 2010). The feed water was stored in 13 L containers at room temperature and replaced weekly. Using conservative tracers, the hydraulic retention time (HRT) in the columns was determined to be 5–6 days (data not shown). Column effluents were sampled 6 days after starting using a new feed solution to account for the HRT in the column system and to provide corresponding sample sets.

Ozonation of feed waters was conducted weekly at room temperature in a 13-L semi-batch stirred tank reactor. Gaseous ozone was produced from pure oxygen using a Modular 8 HC generator (Wedeco, Germany). Ozone concentration of applied and off-gas, dissolved ozone and gas flow rate were measured continuously using BMT 964 and BMT 961 TPC (BMT, Germany), Orbisphere 31331.1 (Orbisphere, Germany) and 807 MFM O2 (Bürkert, Germany) probes, respectively. In order to set up a complete mass balance, all off-gas ozone was stripped with pure oxygen before sampling. Operating conditions and prevailing redox conditions of column experiments are summarized in Table 1.

Description of recirculating column experiments

In addition, bromate removal under anoxic conditions was evaluated in four small-scale recirculating columns (l = 20 cm, d = 5 cm) filled with aquifer material as inoculum collected from Lake Tegel, Berlin (19.5 m depth). The column set-up is exemplarily shown for one column in Figure 1. An initial experiment was conducted with tap water spiked with 10 mg/L lactose-peptone as a biodegradable organic carbon source and 50 µg/L bromate. In addition, 5 mg/L nitrate-N were added in two of the four columns to evaluate the effect of nitrate as a competing

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Table 1 | Description and redox conditions of long-term column experiments (all columns were operated with secondary effluent from Berlin-Ruhleben WWTP)

	SC 1 (reference)	SC 2 (ozonated)	SC 3 (ozonated and anoxic)	SC 4 (oxic)
Pretreatment	–	ozonation with ≈0.7 mg/mg DOC	ozonation with 0.7–1 mg/mg DOC with subsequent purging using N_2	H_2O_2 addition
Operation period	≈3.5 years	≈3.5 years	≈10 months	≈10 months
Influent nitrate (mg N/L)	8.6 ± 2.3	8.5 ± 2.6	8.7 ± 2.2	n.a.
Nitrate removal (mg N/L)	0.7	<0.2	0.9	None
Effluent DO (mg/L)	<0.5	>1.0	<0.5	>1.0
Prevailing redox conditions	Suboxic	Mainly oxic	Anoxic	Oxic

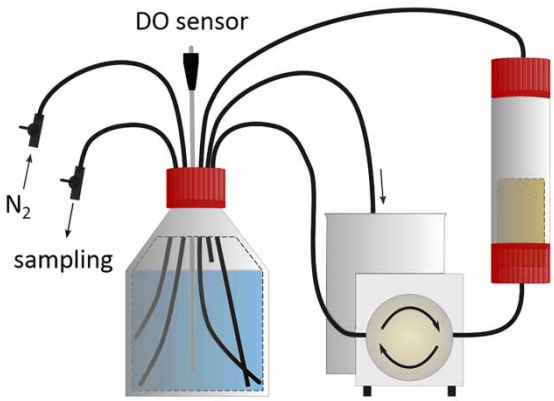


Figure 1 | Set-up of small-scale anoxic column systems.

electron acceptor. For the second experiment, surface water from Lake Tegel was ozonated with 0.9 mg O_3/mg DOC and spiked with different amounts of bromate (50 µg/L and 1 mg/L). Experiments were repeated by refilling reservoirs with 1 L freshly prepared solutions. Forty mL samples were taken by purging nitrogen gas into the reservoir.

Analytical methods

Dissolved oxygen (DO) concentrations and temperature were measured directly after sampling using a WTW Oxi 330 Oximeter (WTW, Germany). Samples for analysis of bulk parameters were filtered through a 0.45 µm cellulose nitrate filter (Sartorius, Germany). DOC was measured using a vario TOC analyzer (Elementar, Germany). Analysis of UV absorbance at 254 nm was conducted on a Lambda 12 photometer (Perkin Elmer, Germany). Nitrate and bromide concentrations were determined by ion chromatography on a Dionex ICS 1100 (Thermo Scientific, Germany).

For measurement of bromate, the liquid chromatography–mass spectrometry (LC/MS) method described by Snyder *et al.* (2005) was adapted. Product ions with m/z 110.9 and 112.9 were detected as quantifier and qualifier ions, respectively. Ninety-eight percent ^{18}O-labeled potassium bromate with a chemical purity of 90–95% (LGC Standards, Germany) was added as an internal standard and the method quantification limit (LOQ) was 0.5 µg/L.

RESULTS

Fate of bulk organic carbon

Results from weekly monitoring of DOC concentrations in secondary effluent and column effluents are shown in Figure 2. A high variation of effluent DOC concentrations from less than 10 to 14 mg/L was observed, which is likely due to diurnal and seasonal variations, but also sampling (as grab samples) and analytical inaccuracies might play a role. In the reference sand column, effluent DOC was removed by 19 ± 8% with a HRT of 5–6 days. Complete consumption of oxygen and additional removal of nitrate in this column system suggested biodegradation as the main removal mechanism. In addition, DOC removal was less efficient during column maturation, ruling out any significant removal contribution by adsorption.

Ozonation of secondary effluent did not significantly change DOC concentrations, which is in line with previous studies (Drewes & Jekel 1998; Hübner *et al.* 2014).

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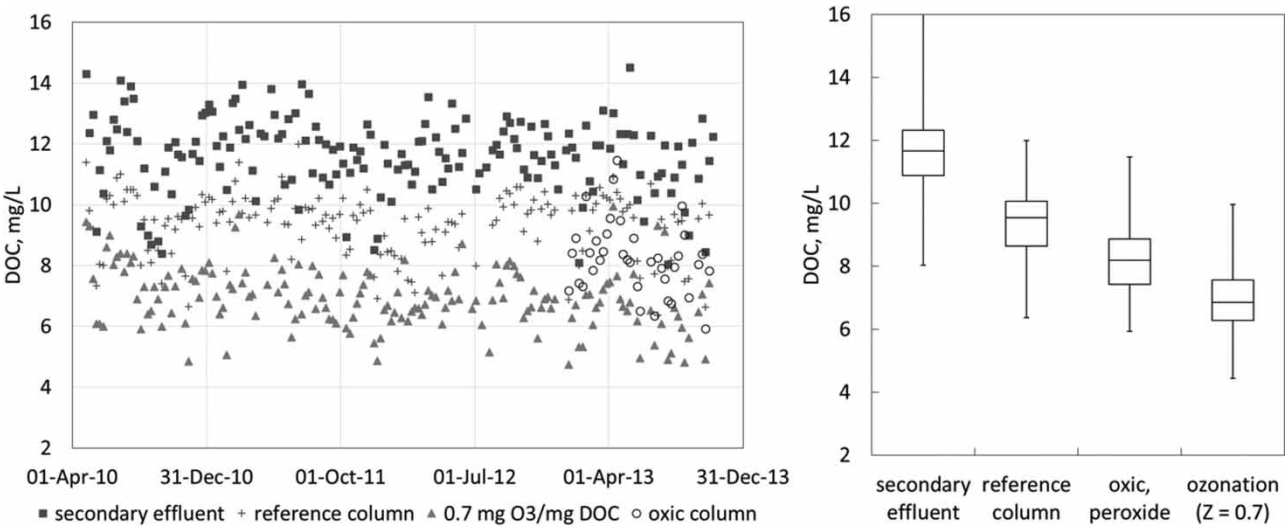


Figure 2 | Monitoring of DOC concentrations in column influent and effluents during long-term column studies (whiskers indicate minimum and maximum concentrations; Z = 0.7 represents a specific ozone consumption of 0.7 mg/mg DOC).

Pretreatment, however, resulted in an increased DOC removal of 40 ± 7% during subsequent passage through sand columns. These results demonstrate that ozonation can strongly enhance biodegradation of organic carbon during subsequent infiltration.

The reasons for the improved DOC removal might be the transformation of DOC due to partial oxidation resulting in higher amenability to biodegradation, and the supply of additional dissolved oxygen in solution favouring aerobic processes. In the reference column without ozonation all oxygen was consumed during infiltration. In addition, analyses of nitrate revealed a removal of approximately 0.7 mg-N/L, indicating the presence of anoxic processes within the column. Redox conditions in the columns fed with ozonated effluent were highly variable due to the ozonation performed weekly. Dissolved oxygen in the feed water container decreased from more than 25 mg/L after ozonation to less than 10 mg/L after 1 week. This fluctuation resulted in partially anoxic conditions in column effluents. Furthermore, changing feed water DO concentrations precluded deriving a correlation between the use of electron acceptors and DOC removal. However, sampling of effluent from column SC 2 for DOC analyses was always scheduled after a completely oxic column passage (as indicated in Table 1). In order to assess the effect of redox conditions on DOC removal, oxic conditions without ozonation were established in column SC4 (by the addition of H_2O_2 to the feed water). Simulated MAR under oxic conditions in SC4 resulted in DOC removal of 27 ± 9% (Figure 2), demonstrating that both the formation of easily degradable DOC and the establishment of oxic redox conditions play an important role in the increased biodegradation of organic carbon after ozonation.

Bromate formation and degradation in long-term column experiments

Bromate formation by ozonation with specific ozone consumption up to 0.9–1.0 mg O_3/mg DOC did not exceed the limit of 10 μg/L as promulgated by the EU Drinking Water Directive (Figure 3). Applying higher ozone dosages resulted in a strong increase in bromate levels. In these cases, especially with respect to water reuse applications, measures for bromate control or removal need to be considered. However, the mechanism for bromate formation during ozonation highly depends on the feed water quality and the transfer of these results to other sites should be used with caution.

Monitoring of bromate concentration in column influents is illustrated in Figure 4. An average bromate formation of 2.9 ± 1.9 μg/L was observed with a specific

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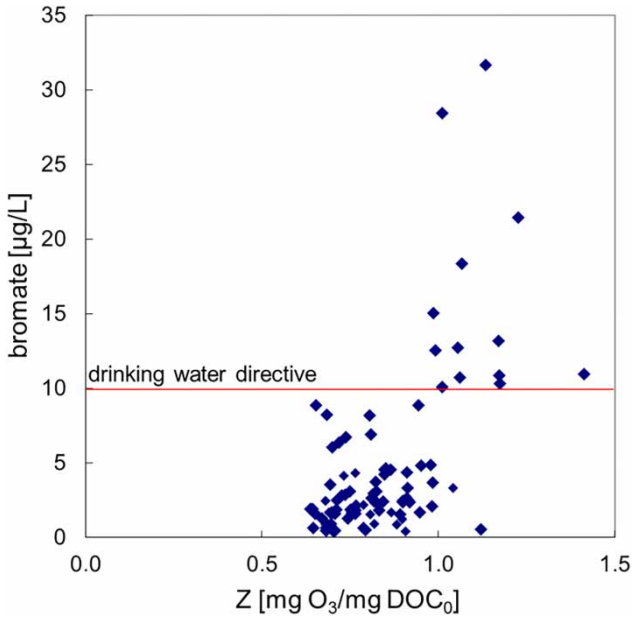


Figure 3 | Bromate formation during ozonation of secondary effluent from the Berlin-Ruhleben WWTP as a function of specific ozone consumption.

ozone consumption of approximately 0.7 mg/mg DOC (feed water for the oxic column). An increase in specific ozone consumption in the second experimental phase resulted in higher bromate levels (feed water for the anoxic column). The high variation in bromate formation is mostly due to changes in the adjusted ozone consumption (consumed ozone depends on ozone dosage and water quality parameters such as DOC, and nitrite) but also weekly changes in water quality might play a role.

As expected, bromate was not efficiently removed during oxic infiltration. Whereas low initial bromate concentrations (2.9 ± 1.9 µg/L) slightly decreased, no significant reduction was observed during the spiking experiment with 100 µg/L bromate. In contrast, the operation of anoxic columns resulted in consistent partial bromate removal of approximately 30% for different initial concentrations. In the same column, a slight but significant removal of nitrate (see values reported in Table 1) suggested the prevalence of anoxic processes within the column. These results indicate simultaneous consumption of bromate and nitrate as electron acceptors.

In combined ozone/MAR systems, however, infiltration of ozonated water occurs under highly oxic conditions and establishment of anoxic conditions would only occur where additional oxygen demand is provided (e.g. percolation through organic rich sediment layers). Therefore, a mitigation of the bromate by-product is not expected to occur during subsequent soil aquifer treatment, and measures for bromate control must be applied during ozonation dependent upon water quality requirements of the desired water reuse practice.

Bromate removal in small-scale anoxic columns

Additional anoxic small-scale column studies were conducted to simulate a worst-case scenario representing a de facto potable reuse situation where secondary treated

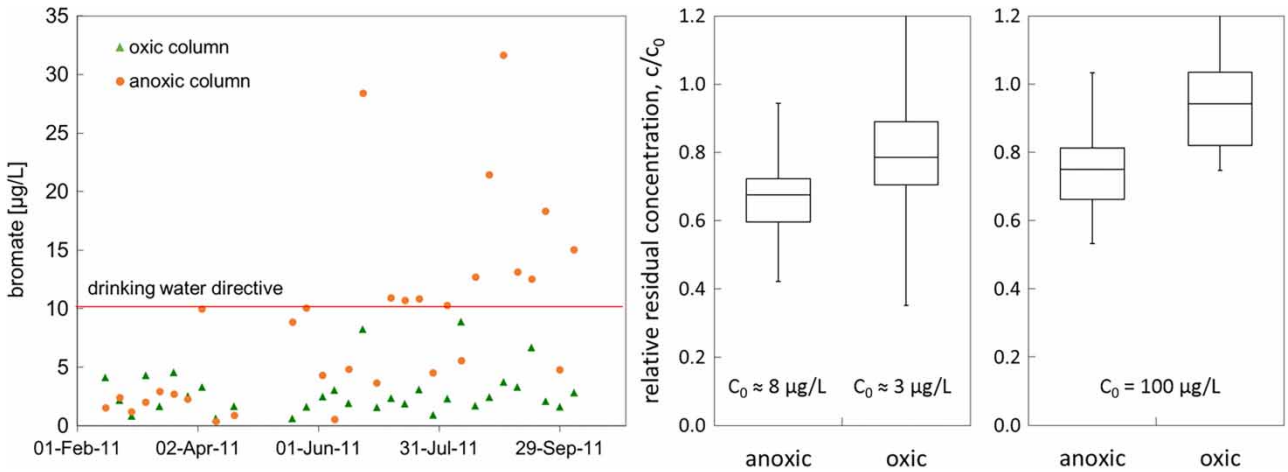


Figure 4 | Monitoring of column feed bromate concentration (after ozonation) and bromate removal in oxic and anoxic sand columns (right: data from spiking experiment with 100 µg/L bromate).

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effluent after ozone and subsequent bank filtration treatment is used for drinking water production. Data from nitrate and bromate analyses in the first experiment are shown in Figure 5. Results from all columns indicate simultaneous consumption of nitrate and bromate during infiltration. In one column with high nitrate concentrations, significantly reduced efficiency of denitrification and bromate removal was observed. Monitoring of DO concentrations in the feed water container (compare the set-up in Figure 1) indicated inhibition of anoxic processes by intrusion of small amounts of dissolved oxygen concentrations in that column system. In the other three columns (one column with high and two columns with low nitrate level), an effect of nitrate concentration on the removal of bromate was not observed.

Results from the second experiment with ozonated water from Lake Tegel and different spiking levels for bromate are shown in Figure 6. As already observed in the first experiment, bromate and nitrate removal was reduced in one column by intrusion of dissolved oxygen. The kinetics of bromate reduction in the other columns were similar, which indicates that the initial bromate concentration did not affect its removal. The results from nitrate measurements confirm previous observations that bromate and nitrate are simultaneously consumed as electron acceptors. Bromide concentration in the spiking experiment with

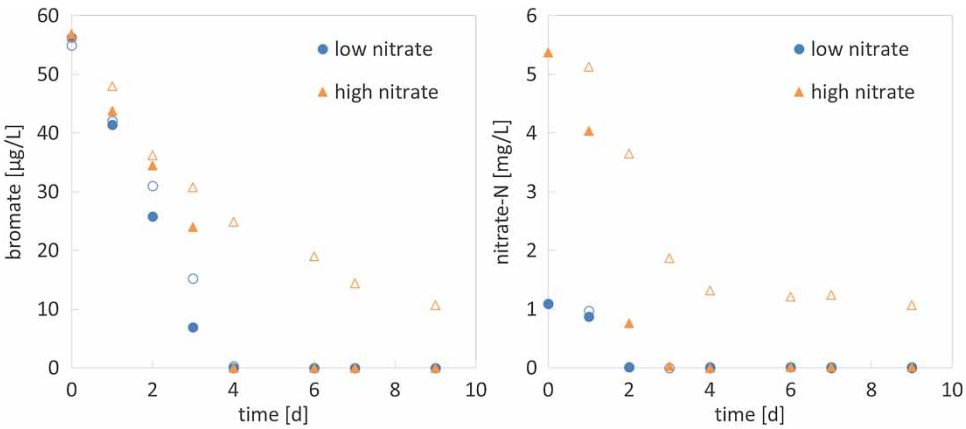


Figure 5 | Bromate removal at different nitrate level (all experiments conducted with 10 mg/L lactose-peptone feed solution, results from duplicate columns are indicated by empty and full signs).

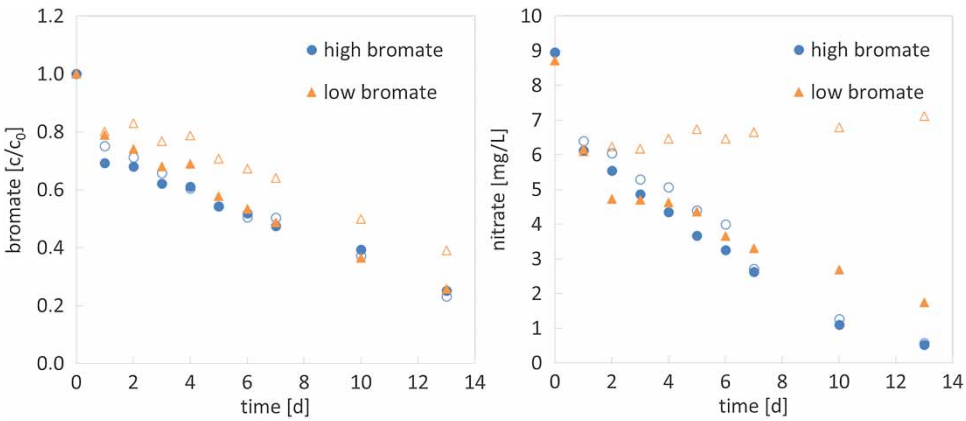


Figure 6 | Nitrate and bromate measurements in small-scale columns with different initial bromate concentration (red: 50 µg/L, blue: 1 mg/L; experiments with preozonation of Lake Tegel water at 0.9 mg/mg DOC). The full colour version of this figure is available in the online version of this paper: http://dx.doi.org/10.2166/wrd.2015.222.

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1 mg/L bromate concentration increased from 120 to 440 μg/L in the columns with intrusion of DO and to 480 μg/L in the fully anoxic column, accounting for a conversion of approximately 70% of reduced bromate-Br into bromide.

In general, faster removal of bromate and nitrate was observed in the first experiment using tap water as the water matrix. A possible explanation is the better availability of organic carbon from lactose-peptone. Unfortunately, a more detailed evaluation of data from DOC analyses was not possible due to carbon leaching from freshly introduced aquifer material.

CONCLUSIONS

The reactions of ozone with effluent organic matter resulted in an increase in microbial degradation of organic carbon during subsequent biological treatment systems. The results from long-term column studies confirm the strong effect of pre-ozonation on DOC removal in MAR systems, increasing removal efficiencies from approximately 20% without pre-treatment to 40% after ozonation. The results from additional oxic column experiments demonstrated that improved DOC removal after ozonation is likely due to two effects, the transformation of DOC into better biodegradable compounds, and the supply of dissolved oxygen to maintain aerobic degradation processes.

Bromate formation during ozonation of secondary effluent from the Berlin-Ruhleben WWTP was constantly below 10 μg/L, the limit of the EU Drinking Water Directive, using a specific ozone consumption of 0.7 mg/mg DOC. Only ozonation at a specific dose of 0.9–1.0 mg O_3/mg DOC resulted in exceedance of the bromate limit for drinking water treatment. Controlled column studies simulating MAR revealed inefficient bromate removal under oxic conditions. The introduction of already small amounts of dissolved oxygen into the porous media strongly affected the reduction of bromate (and nitrate) during subsurface treatment. During anoxic infiltration, bromate can be used by bacteria for anaerobic growth, and it is reduced to bromide. In contrast to several previous studies, which reported strong inhibition of bromate removal in the presence of nitrate (Hijnen *et al.* 1999; Matos *et al.* 2008), simultaneous reduction of bromate and nitrate was confirmed in several independent experiments. This transformation might be explained by higher microbial diversity in groundwater recharge systems, since not all denitrifying bacteria are capable of bromate reduction and bromate specific reaction mechanisms likely exist (Van Ginkel *et al.* 2005; Davidson *et al.* 2011).

These results demonstrate that an accumulation of bromate in partially closed urban water cycles including prevailing anoxic bank filtration conditions is not to be expected. However, groundwater recharge and soil aquifer treatment systems after ozonation usually result in highly oxic subsurface conditions, and anoxic conditions might only occur in infiltration systems with efficient aeration (stripping of excess oxygen) and/or percolation through organic-rich sediment layers. In these situations, MAR would not provide efficient bromate mitigation and bromate levels in reclaimed water would need to be controlled upstream during ozonation.

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REFERENCES

Clara, M., Strenn, B., Gans, O., Martinez, E., Kreuzinger, N. & Kroiss, H. 2005 Removal of selected pharmaceuticals, fragrances and endocrine disrupting compounds in a membrane bioreactor and conventional wastewater treatment plants. *Water Res.* **39** (19), 4797–4807.

Davidson, A. N., Chee-Sanford, J., Lai, H. Y. M., Ho, C.-H., Klenzendorf, J. B. & Kirisits, M. J. 2011 Characterization of bromate-reducing bacterial isolates and their potential for drinking water treatment. *Water Res.* **45** (18), 6051–6062.

Downing, L. S. & Nerenberg, R. 2007 Kinetics of microbial bromate reduction in a hydrogen-oxidizing, denitrifying biofilm reactor. *Biotechnol. Bioeng.* **98** (3), 543–550.

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Drewes, J. E. & Jekel, M. 1998 Behavior of DOC and AOX using advanced treated wastewater for groundwater recharge. *Water Res.* **32** (10), 3125–3133.

Drewes, J. E., Heberer, T., Rauch, T. & Reddersen, K. 2003 Fate of pharmaceuticals during ground water recharge. *Ground Water Monit. Remed.* **23**, 64–72.

Drewes, J. E., Hoppe, C. & Jennings, T. 2006 Fate and transport of N-nitrosamines under conditions simulating full-scale groundwater recharge operations. *Water Environ. Res.* **78** (13), 2466–2473.

Grünheid, S., Amy, G. & Jekel, M. 2005 Removal of bulk dissolved organic carbon (DOC) and trace organic compounds by bank filtration and artificial recharge. *Water Res.* **39** (14), 3219–3228.

Hijnen, W. A. M., Jong, R. & Van Der Kooij, D. 1999 Bromate removal in a denitrifying bioreactor used in water treatment. *Water Res.* **33** (4), 1049–1053.

Hollender, J., Zimmermann, S. G., Koepke, S., Krauss, M., McArdell, C. S., Ort, C., Singer, H., von Gunten, U. & Siegrist, H. 2009 Elimination of organic micropollutants in a municipal wastewater treatment plant upgraded with a full-scale post-ozonation followed by sand filtration. *Environ. Sci. Technol.* **43** (20), 7862–7869.

Hoppe-Jones, C., Oldham, G. & Drewes, J. E. 2010 Attenuation of total organic carbon and unregulated trace organic chemicals in US riverbank filtration systems. *Water Res.* **44** (15), 4643–4659.

Hübner, U., Miehe, U. & Jekel, M. 2012 Optimized removal of dissolved organic carbon and trace organic contaminants during combined ozonation and artificial groundwater recharge. *Water Res.* **46** (18), 6059–6068.

Hübner, U., Seiwert, B., Reemtsma, T. & Jekel, M. 2014 Ozonation products of carbamazepine and their removal from secondary effluents by soil aquifer treatment – Indications from column experiments. *Water Res.* **49** (1), 34–43.

Joss, A., Zabczynski, S., Göbel, A., Hoffmann, B., Löffler, D., McArdell, C. S., Ternes, T. A., Thomsen, A. & Siegrist, H. 2006 Biological degradation of pharmaceuticals in municipal wastewater treatment: proposing a classification scheme. *Water Res.* **40** (8), 1686–1696.

Kirisits, M. J., Snoeyink, V. L., Inan, H., Chee-Sanford, J. C., Raskin, L. & Brown, J. C. 2001 Water quality factors affecting bromate reduction in biologically active carbon filters. *Water Res.* **35** (4), 891–900.

Matos, C. T., Velizarov, S., Reis, M. A. M. & Crespo, J. G. 2008 Removal of bromate from drinking water using the ion exchange membrane bioreactor concept. *Environ. Sci. Technol.* **42** (20), 7702–7708.

Reemtsma, T., Weiss, S., Mueller, J., Petrovic, M., Gonzalez, S., Barcelo, D., Ventura, F. & Knepper, T. P. 2006 Polar pollutants entry into the water cycle by municipal wastewater: a European perspective. *Environ. Sci. Technol.* **40** (17), 5451–5458.

Scheurer, M., Godejohann, M., Wick, A., Happel, O., Ternes, T., Brauch, H. J., Ruck, W. & Lange, F. 2012 Structural elucidation of main ozonation products of the artificial sweeteners cyclamate and acesulfame. *Environ. Sci. Pollut. Res.* **19** (4), 1107–1118.

Schumacher, J., Pi, Y. Z. & Jekel, M. 2004 Ozonation of persistent DOC in municipal WWTP effluent for groundwater recharge. *Water Sci. Technol.* **49** (4), 305–310.

Schwarzenbach, R. P., Escher, B. I., Fenner, K., Hofstetter, T. B., Johnson, C. A., Von Gunten, U. & Wehrli, B. 2006 The challenge of micropollutants in aquatic systems. *Science* **313** (5790), 1072–1077.

Snyder, S. A., Vanderford, B. J. & Rexing, D. J. 2005 Trace analysis of bromate, chlorate, iodate, and perchlorate in natural and bottled waters. *Environ. Sci. Technol.* **39** (12), 4586–4593.

Ternes, T. A., Meisenheimer, M., McDowell, D., Sacher, F., Brauch, H.-J., Haist-Gulde, B., Preuss, G., Wilme, U. & Zulei-Seibert, N. 2002 Removal of pharmaceuticals during drinking water treatment. *Environ. Sci. Technol.* **36** (17), 3855–3863.

Tyler, C. R., Spary, C., Gibson, R., Santos, E. M., Shears, J. & Hill, E. M. 2005 Accounting for differences in estrogenic responses in rainbow trout (Oncorhynchus mykiss: Salmonidae) and roach (Rutilus rutilus: Cyprinidae) exposed to effluents from wastewater treatment works. *Environ. Sci. Technol.* **39** (8), 2599–2607.

Van Ginkel, C. G., Van Haperen, A. M. & Van Der Togt, B. 2005 Reduction of bromate to bromide coupled to acetate oxidation by anaerobic mixed microbial cultures. *Water Res.* **39** (1), 59–64.

Yoon, M. K., Drewes, J. E. & Amy, G. L. 2013 Fate of bulk and trace organics during a simulated aquifer recharge and recovery (ARR)-ozone hybrid process. *Chemosphere* **93** (9), 2055–2062.

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