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ABSTRACT Routine quality monitoring for fecal indicators after ozonation at the river-lake waterworks Weesperkarspel of Amsterdam Water Supply (AWS) show large variation in inactivation. The influence of the high DOC in the water on the inactivation efficiency was investigated. Results showed a higher inactivation of *Clostridium perfringens* in the AWS water than in a water with low DOC at the same CT conditions. The contribution of the gas feed chamber to the overall inactivation of *C. perfringens* was high in the AWS water and was reduced after DOC reduction with GAC. This result may alter the current CT concept of the process. Further research will be focused on the ozone dosage strategy and control related to the required CT for inactivation and the production of by-products.

KEY WORDS Ozone; Amsterdam Water Supply; *Clostridium perfringens*; *Bacillus subtilis*; *Escherichia coli*; Disinfection Kinetics

Drinking water is produced from surface water at the river-lake waterworks Weesperkarspel of the Amsterdam Water Supply (AWS). One of the treatment barriers against pathogenic microorganisms is ozonation. The current process conditions are optimized for disinfection, cost, and by-products formation (bromate and AOC). Routine water quality monitoring with fecal indicator bacteria (thermotolerant coliforms and spores of sulfite-reducing clostridia) showed large variation disinfection performance (Hijnen et al., 2001). Due to the slow sand filters as additional barriers the suboptimal ozone performance has not led to noncompliance with the standards for indicator bacteria in the drinking water. Nevertheless, AWS wants to operate a robust ozone disinfection and has begun a project to optimize this process.

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Water quality is a significant factor in predicting the disinfection efficacy (Haas et al., 1996). One hypothesis for the poor disinfection performance of the ozonation is the relatively high dissolved organic carbon (DOC) concentration in the source water. The DOC of 5–6 mg/L (UV extinction of 12–15 m^{-1}) results in a high ozone demand and decay rate, especially in summer. A lab-scale study is carried out to determine the effect of the DOC reduction in the water on the inactivation capacity of ozonation. A continuous flow system with gas feed and contact chambers is used to simulate full-scale conditions. *Clostridium perfringens* was used as a model organism to verify the disinfection performances under different conditions. This indicator bacterium and also the group of sulfite-reducing clostridia (SSRC) have been proposed as surrogate parameters to monitor the treatment efficiency for the removal of persistent pathogenic viruses and protozoa (Hijnen et al., 1997, 2000a, 2002; Payment and Franco, 1993). Simultaneously, the inactivation of *Escherichia coli* and *Bacillus subtilis* was determined as microorganisms with a high and low susceptibility for ozone, respectively. The latter is representative of the aerobic spores which have been proposed as a surrogate for the removal of persistent pathogens as well (Rice et al., 1996).

MATERIALS AND METHODS

Experimental setup

Q1 Disinfection experiments were performed in the source water of AWS with and without a reduced DOC content. The inactivation of spores of *C. perfringens* was determined in a lab-scale continuous flow ozone system at a constant water temperature of 10°C. This experimental setup has been used (Hijnen et al., 2002) for experiments in the water of Water Company Europoort (WBE) with a low DOC content (Table I).

In the first experiment, DOC reduction in the AWS water was achieved by enhanced coagulation followed by filtration (AWS-EC). The pH of source water was reduced to 4.5, and 6 mg/L flocculant were added before plane sedimentation. After sedimentation, the coagulated water was filtered through a rapid sand filter (sand 0.7–1.4 mm). This pretreatment resulted in a DOC reduction of 1 mg/L DOC (Table I).

TABLE I Major Characteristics of the Waters Used in the Study

Water type	AWS	AWS-EC	AWS-GAC1/2	WBE
DOC (mg/L)	5.4; 5.5	4.3	4.4; 3.7	2.1
pH	7.7; 7.6	7.4	7.7; 7.8	7.4
Turbidity (Fte)	0.16; 0.18	0.85	nd	0.08
Fe (mg/L)	nd[a]	0.5	nd	0.018

[a] nd, not determined.

The second series of experiments were carried out with the AWS water filtered over granular activated carbon (GAC) after 7,680 bed volumes. The water was sampled after a contact time of 60 (AWS-GAC1) and 120 min (AWS-GAC2), resulting in a DOC reduction of 1.1 and 1.8 mg/L, respectively (Table I).

Ozone System

The Kiwa continuous flow system (Figure 1) is a lab-scale replicate of the full-scale WBE ozonation process. Technical information about the lab-scale system and the two full-scale processes of AWS and WBE is summarized in Table II.

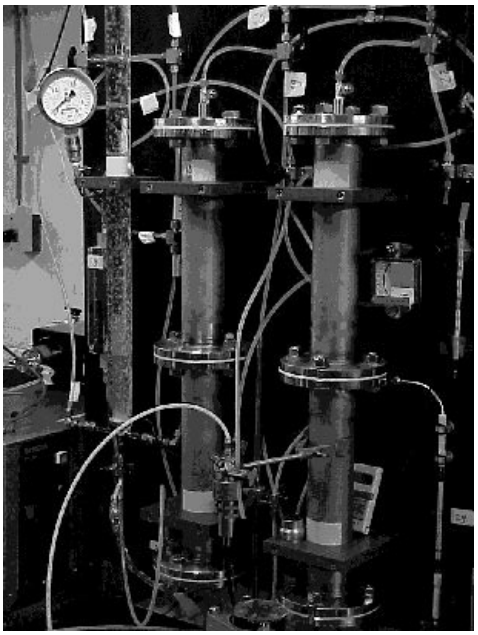

FIGURE 1 The lab-scale ozone system.

TABLE II Characteristics of the Ozone Systems

Characteristics	Laboratorium	WBE	AWS
Volume chambers (m^3)	0.0014[a]; 0.0026[b]	4.1[a]; 4.1[b]	396[a]; 1,472[b]
Water flow (m^3/h)	0.042	100	3,400
Gas flow (m^3/h)	0.012	26	331[c]
Transfer efficiency (%)	90–93	95	95–98
T_{10}/HRT (min/min)	0.75	0.72	0.60

[a] Gas-feed counter-current chambers (GCC).
[b] Contact chambers (CC).
[c] Constant gas/water ratio (constant ozone dose).

Ozone Concentrations and CT Calculation

Ozone in water was determined by the Indigo method (Bader and Hoigne, 1982). The CT value was calculated from the average ozone concentration (mg/L) and the average contact time T (min) in each of the successive chambers (gas feed (GC) and contactors (CC)). The average ozone concentrations were calculated from the in- and outgoing concentration $(C_{O_3,in} + C_{O_3,out})/2$. To incorporate the influence of the contact time distribution in the system T_{10} was used, defined as the contact time in which 90% of the incoming water has passed the chamber (Von Huben, 1991). T_{10} values of the different systems were determined with tracer tests (Cl^-) and presented as ratio of the hydraulic retention time (HRT; Table II).

Microorganisms and Microbiological Analysis

The model organism *C. perfringens* was an isolate from a patient who had from diarrhea (strain D10). The methods for preparing a stock solution with high spore concentrations and inoculation of the water before ozonation were described earlier (Hijnen et al., 2002). Spore concentration in the water prior to ozonation was 10^3–10^4 CFU/mL.

Samples before and after ozonation were collected in sterile bottles with sterilized thiosulfate solution (2 mL/L of a 30 g/L solution) as a quenching agent. *C. perfringens* D10 and SSRC concentrations in water were determined on sulfite-iron agar by the method previously described (Hijnen et al., 1997). The 1 or 0.1 mL pasteurized samples (30 min at 70°C) were inoculated directly in the liquid agar medium, with or without further dilution in 9 mL of sterile drinking water. The standard membrane filtration method was used for enumerating SSRC in the feed water of the full-scale ozonation and the MF sampling technique (Hijnen et al., 2000b) was used for SSRC enumeration in large volumes (100 L) after this process.

Freeze-dried spores of *B. subtilis* (ATTC6633) were suspended in autoclaved water before inoculation in the test water and enumerated on plate count agar (Difco 247940) incubated at 37±(1°C for 24 h. *E.coli* (WR1) was cultured overnight in liquid, nutrient-rich medium; then washed, centrifuged, and diluted in autoclaved water before inoculation in the test water. For enumeration, ISO 9308-1 with lauryl sulfate agar (Oxoid MM615) was used.

MEASUREMENTS AND RESULTS

Inactivation of *C. perfringens*

The ozone dose to obtain a CT_{10} and inactivation of *C. perfringens* in the AWS water was approximately 1.7 mg/L (Table III). Both CT_{10} and inactivation increased with increasing ozone dose. DOC reduction of 1.1 mg/L by enhanced coagulation (EC) did not yield a higher CT_{10}. Moreover, the level of inactivation was lower than in high DOC water at comparable CT_{10}. This probably was caused by the high turbidity and Fe concentration in the EC water (Table I). This will have masked an effect of DOC reduction.

DOC reduction with 1.1 and 1.8 mg/L by GAC filtration (AWS-GAC1 and AWS-GAC2, respectively) yielded higher CT_{10} values and more inactivation of *C. perfringens* was achieved at the same transferred ozone dose (Table III). Furthermore, inactivation of *C. perfringens* spores in the AWS-GAC water seemed

TABLE III The Inactivation of Spores of *C. perfringens* (log_{10} ± range of duplicate samples) at 10°C Determined in the AWS Water, the AWS Water Treated for DOC Reduction and the WBE Water Related to the Transferred Ozone Dose (mg/L) and the CT10 (mg/L)*min

AWS water[a]			AWS: DOC reduced			WBE water		
Dose	CT_{10}	Inact.	Dose	CT_{10}	Inact.	Dose	CT_{10}	Inact.
1.65	0.11	0.6 ± 0.05		AWS-EC		0.90	0.74	0.5 ± 0.00
1.79	0.46	0.4 ± 0.05	1.07	0.30	0.0	1.03	1.10	0.7 ± 0.00
1.84	0.61	0.1 ± 0.20	1.41	0.55	0.0	1.23	1.53	0.8 ± 0.05
2.28	1.48	1.7 ± 0.10	2.86	1.46	0.2 ± 0.05	1.40	1.98	1.0 ± 0.05
2.32	1.29	1.6 ± 0.20	4.20	3.64	2.2 ± 0.20	1.54	2.25	1.3 ± 0.30
2.61	0.69	0.7 ± 0.20		AWS-GAC1		1.79	2.87	1.5 ± 0.15
3.12	2.96	2.2 ± 0.30	1.58	0.96	1.6 ± 0.00	2.16	4.11	2.0 ± 0.10
3.04	3.20	2.5 ± 030	2.65	3.73	2.6 ± 0.00	3.04	6.31	2.8 ± 0.30
3.97	6.30	3.5 ± 0.15	4.20	9.58	3.9 ± 0.30			
4.12	6.44	2.7 ± 0.30		AWS-GAC2				
4.35	3.13	2.6 ± 0.20	1.68	0.92	1.2 ± 0.00			
			2.79	4.30	2.7 ± 0.10			
			4.41	10.87	4.2 ± 0.40			

[a]Two batches of water.

to be more efficient than in the low DOC water of WBE; more inactivation was observed in the AWS-GAC at comparable CT_{10}.

Inactivation of Environmental SSRC

Under full-scale conditions, the inactivation of environmental spores of sulfite-reducing clostridia (SSRC) was determined and compared with the lab-scale results of *C. perfringens* spores. Average inactivation of SSRC in summer (17.5°C) at a constant ozone dose of 1.7 mg/L was 1.5 log_{10} and showed a large variation (0.2, 0.4, 1.95, 2.6, 2.3). In winter (5.3°C), SSRC were inactivated with 0.8 log_{10} (0.4, 0.7, 0.7, 1.2) at a constant ozone dose of 2.2 mg/L.

These results show that the inactivation of SSRC under full-scale conditions was on the same order of magnitude as *C. perfringens* spore inactivation in lab-scale experiments. The observed differences in inactivation of the environmental SSRC and lab-cultured *C. perfringens* were probably caused by the significantly higher and lower temperature in the full-scale system.

Inactivation of *E.coli* and *B. subtilis* Spores

In some experiments, *E. coli* or *B. subtilis* were inoculated in the water simultaneously with *C. perfringens* spores. *E. coli* was much more efficiently inactivated by ozone than *C. perfringens*. At a transferred ozone dose of 1.84 mg/L (CT_{10}=0.61 (mg/L)*min) *C. perfringens* was inactivated with 0.1 log (Table III) and *E. coli* with 3.3 log (Table IV). As was observed for *C. perfringens*, the efficiency of inactivation of *E. coli* in AWS water with reduced DOC content by enhanced coagulation did not increase but was reduced instead (Table IV).

The effect of ozone on the *Bacillus* spores was significantly lower than the effect on *Clostridium* spores. Up to a CT_{10} value of 4 (mg/L)*min, no inactivation was observed. Only in the AWS-GAC water and a dose of 4.20 and 4.41 mg/L was an inactivation of 0.7 and 1.0 log determined, respectively. Under these conditions, *C. perfringens* was inactivated with 3.9 and 4.2 log, respectively.

TABLE IV The Inactivation (log_{10}) of *E. coli* at 10°C Related to the Transferred Ozone Dose (mg/L) and the CT_{10} Value ((mg/L)*min)

AWS			AWS-EC		
Dose	CT_{10}	Inact.	Dose	CT_{10}	Inact.
1.84	0.61	3.3	1.07	0.30	0.3
2.32	1.29	3.7	1.41	0.55	0.8
3.12	2.96	>4.3	2.86	1.46	3.3
4.12	6.44	>4.3	4.20	3.64	>4.5

DISCUSSION

Inactivation Kinetics

Disinfection is the reduction of the concentration viable microorganisms N due to the exposure to a concentration disinfectant C during a specific contact time T. The relationship between the CT value ((mg/L)*min) and the observed inactivation is commonly described by the first-order disinfection models of Chick (1908) and Watson (1908). As described before (Hijnen et al., 2002) and presented in Figure 2, the disinfection kinetics of *C. perfringens* spores in the WBE water can be described by the Chick model with an inactivation rate constant k of –0.25 ($SD = 0.014$; $p < 0.001$; $n = 1$).

Data in this study showed a nonlinear relation between inactivation and CT_{10} in the AWS water (Figure 2) and the inactivation data in the AWS-GAC water fitted well in this nonlinear relation. The data were fitted with the model proposed by Watson (1908) with parameter n as the dilution constant

$$\ln\left(\frac{N_t}{N}\right) = -kC^nT$$

N is the initial concentration and N_t the concentration after contact time T (min) and k is the inactivation rate constant. The data resulted in a k value of –0.49 and an n value of 0.64 ($p = <0.001$; $R^2 = 0.69$). The plot is presented in Figure 2.

Water Quality and Disinfection

From thier study, Haas et al. (1996) concluded that turbidity and inorganic chemical composition affect inactivation efficiency of ozonation. This study also demonstrated a negative impact of turbidity and residual iron after coagulation and filtration on the inactivation of *C. perfringens* and *E. coli* (Tables III and V). The low levels of inactivation could be attributed to protection of microorganisms attached to colloidal particles in the water. This explanation is consistent with other literature data (LeChevallier, 1981).

Helmer and Finch (1993) described how DOC is an important factor for inactivation of MS2 bacteriophages in natural waters. High DOC concentrations in the water will result in a high instantaneous ozone demand (ID; mg/L) of the water and a high-first order decay rate k_d of ozone in the contactors (Table V). By reducing the DOC in the AWS water by GAC, the ID decreased with ±35%. The decay rate k_d in the AWS-GAC water was ±50% lower than in

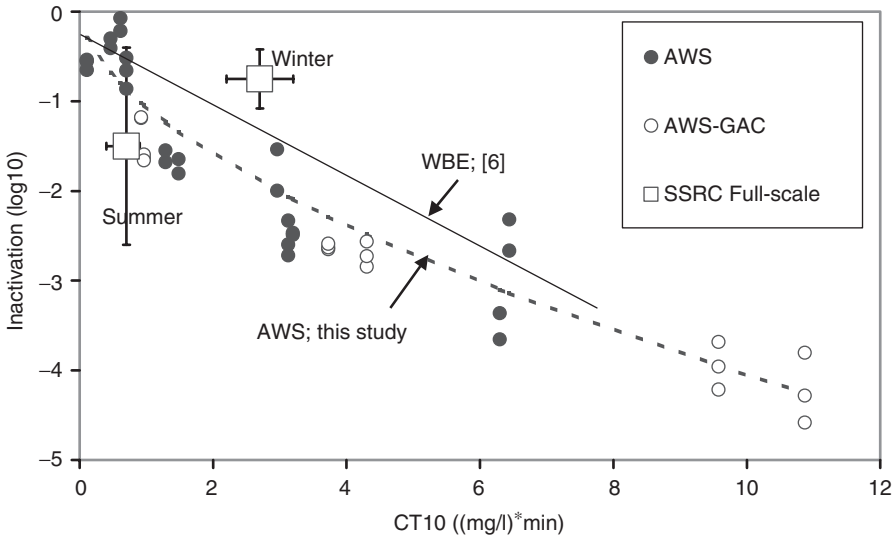

FIGURE 2 The inactivation of spores of *C. perfringens* in the AWS water (●) and in the AWS water with reduced DOC content (AWS-GAC; ○) at 10°C related to the calculated CT_{10} value; the inactivation of environmental SSRC in the full-scale process determined in winter and summer (□; range of data presented by error bars).

TABLE V The Instantaneous Ozone Demand (ID) and the Ozone Decay Rate k_d Determined for the Tested Waters at 10°C

	AWS	AWS-GAC	WBE
DOC (mg/L)	5.4; 5.5	4.4; 3.7	2.1
ID (mg/L)	1.6; 1.9	1.1; 1.2	0.4
kd[a] (min^{-1})	0.44	0.29 ± 0.02	0.11 ± 0.03

[a]Determined at a transferred ozone dose of 2.4–2.8 mg/L.

untreated AWS water, but still three times higher than in the WBE water. Consequently, a higher DOC implicates that a higher ozone dose is needed to achieve a required CT value (Table III).

From their study, Haas et al. (1996) concluded that the disinfectant demand is a function of water quality, but not necessarily a good predictor of the inactivation efficiency. Moreover, Finch et al. (2001) reviewed and analyzed literature data of *Cryptosporidium* inactivation by ozone and concluded that there was no single kinetic model which gave a "best fit" to all data sets. Both conclusions are supported by the results described in this study. Comparison of the inactivation of *C. perfringens* in both waters showed that, due to this difference in disinfection kinetics, inactivation in the AWS water with high DOC was more efficient than in the low DOC water of WBE up to a CT_{10} value of 7 (mg/L)*min (Figure 2).

Variation in Inactivation

The logarithmic kinetic model observed for the AWS water implicates that, at low CT_{10} values, a variation in CT_{10} will have a large effect on the inactivation efficiency. There were data that showed such an effect under full-scale conditions. Inactivation of environmental SSRC by the full-scale ozonation in summer was highly variable at a low ozone dose of 1.7 mg/L and CT_{10} (Figure 2). The CT_{10} of the process is variable due to the applied constant ozone dose (Table II). In winter at 3°C to 5°C and a higher constant dose of 2.2 mg/L and higher $CT_{10,}$ inactivation of SSRC was more stable.

Inactivation During Ozone Transfer

DOC in the water influences the instantaneous ozone demand in the gas feed chamber and the ozone decay in the contactors. To determine the significance of both effects on the inactivation efficiency, the inactivation during ozone transfer in the gas feed chamber and in the contactors was determined separately. The results showed a high level of inactivation of *C. perfringens* in the AWS water during ozone transfer in the gas feed chamber: 80.8 ± 29.6% of the overall inactivation occurred in this chamber, while only 20–40% (29.6 ± 7.2 %) of the CT_{10} in the AWS and AWS-GAC water was produced in the gas feed chamber (Figure 3).

A higher inactivation efficiency during gas transfer than during contact with dissolved ozone alone was previously reported (Finch and Smith, 1991; Masschelein, 1982; Farooq et al., 1977) and was attributed to a higher inactivation efficiency in the gas-liquid interface (Farooq et al., 1977). The effect was reduced at reduced DOC after GAC filtration: In the AWS-GAC water, inactivation of *C. perfringens* in the gas feed chamber was reduced to 40.2 ± 17.6% of the overall inactivation. Furthermore, it was not observed in the low DOC water of WBE where inactivation of *C. perfringens* in the gas feed chamber was proportional to the inactivation observed in the contactors (Figure 3). This indicates a possible effect of DOC on inactivation during gas transfer.

Similarly high inactivation in the gas feed chamber was observed for *E. coli* suspended in the AWS and the AWS-EC water (Table VI). However, the effect was not observed for *B. subtilis*. Inactivation of the spores occurred almost completely during contact with dissolved ozone in the contactors (Table VI), an observation also described recently in literature (Craik et al., 2002). Nevertheless, these results demonstrate that common practice where the CT in the gas feed chamber is excluded from the overall CT (Von Huben, 1991) may significantly underestimate the overall disinfection capacity of the process under certain conditions for some microorganisms.

Inactivation in the Contactors

The relative low inactivation in the contactors (Figure 3) was due to the low ozone concentrations. At an ozone dose of 2.5 mg/L, ozone concentrations after contactor 1 were <0.1 mg/L; no inactivation

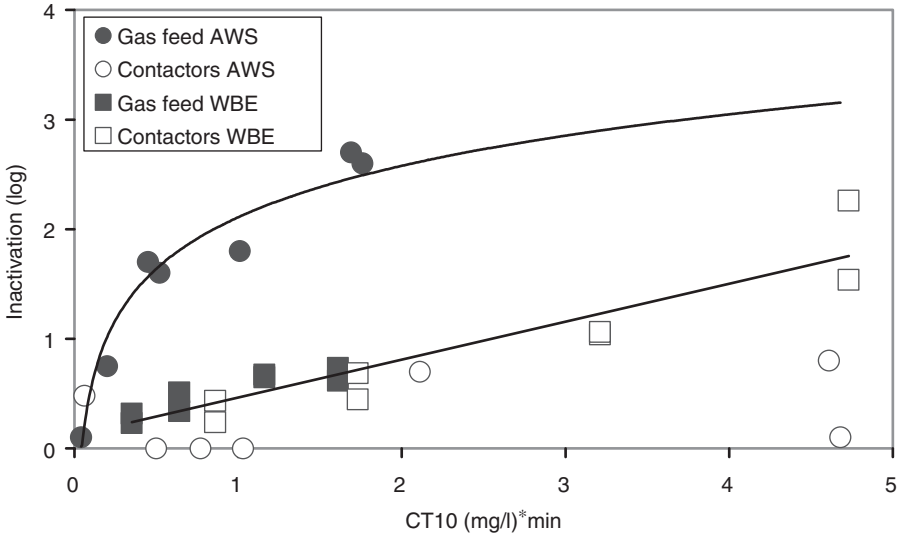

FIGURE 3 The inactivation of spores of *C. perfringens* in the AWS water and the WBE water at 10°C determined in the lab-scale system after the gas feed chamber and the two contactors related to the CT_{10} value were calculated for these separate segments.

TABLE VI The Fraction of Inactivation of *E. coli*, *C. perfringens*, and *B. subtilis* in the Gas Feed Chamber

	Ozone conditions			% Inact. in GC		
Water	Dose	CT_{10}	$\%CT_{10}$ in GC	*E. coli*	*C. Perfringens*	*B. subtilis*
AWS	2.32	1.29	40	97.3	100	nd[a]
AWS-EC	1.41	0.55	16.6	100	Ndis[b]	nd
AWS-GAC1	4.20	9.58	22.5	nd	56.4	14.3
AWS-GAC2	4.41	10.87	21.4	nd	52.4	10

[a]nd, not determined.
[b]Ndis, no disinfection.

occurred in the contactors. In the AWS-GAC water at an ozone dosage of 2.5 mg/L, the ozone concentration after contactor 1 was 0.4 mg/L and *C. perfringens* spores were inactivated with 1.1–2.1 log in these chambers. The effect might be enhanced by heterogeneous susceptibility of microorganisms and low susceptibility of aggregates of microorganisms or protection by encapsulation in particles or colloids.

CT Calculation

It should be noted that the applied CT calculation in this research assumes a linear ozone increase and decrease in gas feed chamber and contactors, respectively. Thus, CT of the countercurrent gas feed chamber and the contactors probably will be overestimated. Due to the difference in instantaneous ozone demand and ozone decay rate observed for the AWS and the WBE water, CT in AWS water probably will be more overestimated than in the low DOC water of WBE.

Assessment of Full-Scale Disinfection Capacity

Environmental SSRC can be used to determine inactivation efficiency of ozonation under full-scale conditions. It is also a potential surrogate for protozoan inactivation. This was concluded from the results of lab-scale studies with *C. perfringens* and of SSRC monitoring under full-scale conditions (Hijnen et al., 2000a). The susceptibility of the lab-cultured *C. perfringens, C. parvum* (animal infectivity; Hijnen et al. (2002)), and environmental

SSRC (Hijnen et al. (2002) and this study) for ozone are on the same order of magnitude. Driedger et al. (2001) concluded that *B. subtilis* is a good surrogate for *C. parvum* at high temperatures, but is more readily inactivated at low temperatures. This study showed that aerobic spores are very resistant against ozone and therefore not appropriate to use under the Dutch full-scale conditions with relatively low CT values to limit bromate production.

CONCLUSIONS

From the results of this study the following conclusions can be drawn:

- A high DOC content in the water before ozonation will result in a high ozone demand, but will not necessarily result in a low inactivation efficiency.
- The observed inactivation kinetics in the AWS water underline the need to apply residual ozone monitoring for ozone dosage control in practice rather than the application of a constant ozone dosage controlled by the production water flow. This adaptation will enhance the stability of inactivation efficiency, especially at low ozone dosages and high temperatures.
- The observed high inactivation efficiency during ozone transfer must be investigated in more detail, especially the conditions under which this occurs. It may alter the current CT concept of the full-scale system, where the CT over the gas feed chamber is not included.
- Spores of sulfite-reducing clostridia, large-volume sampling, and lab-scale studies are useful tools for optimization studies for full-scale ozone disinfection. Aerobic spores are less suitable due to their high resistance against ozone at low CT.
- DOC reduction before ozonation by enhanced coagulation will not increase, but instead will reduce inactivation capacity when turbidity and/or residual iron are insufficiently removed.
- Further research must be initiated to investigate the effect of the presented results on the optimization of the process design and control of the full-scale ozonation. This research will be focused on the ozone dosage strategy and control related to the required CT for inactivation and the production of by-products bromate and AOC.

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