Benefits of Ozone Treatment for Bottled Water

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Abstract

The ozone treatment enhances the water quality of most drinking water in general. However, it is a key and essential treatment for the production of safe, high quality, good tasting, aesthetically pleasing and storage stable bottled water that the consumers have come to expect. The development and adaptation of ozone treatment in the 1970's resolved the troublesome and sometimes embarrassing problems with disinfection and storage stability and set the water bottling industry onto its decades long rapid double digit growth pattern. By the year 2001, the annual sales of bottled water exceeded 5.5 billion gallons and surpassed $ 6 billion dollars in the US. A discussion of the biological and chemical processes involved in the treatment are presented below.

Ozone treatment is the most frequently used disinfection process in water bottling today. With the application of a single ozone treatment step, the water bottler can disinfect the water, the bottling equipment, the bottle, the air above the water and the sealed cap of the bottle, thereby provide a most effective barrier to microbiological contamination for the protection and benefit of the consumer. These are the reasons why most water bottlers rely on ozone treatment to provide a safe, good tasting, aesthetically pleasing and storage stable product. As the ozone treatment became a well accepted, routine part of the water bottling process, many of its other benefits beyond the disinfection have become taken for granted and nearly forgotten. Some water bottlers may not even realize today that the use of ozone also provides benefits, such as, improved taste, elimination of odor, and long 2 years storage stability. These benefits have improved their product substantially making it a good tasting and safe. These are the product quality features that the costumers have grown to expect and enjoy. Since ozone treatment has become such a key process for the water bottlers, they need to stay current with the advances in the technology, and the improvements in the ozone treatment process. In addition they must follow and review the regulatory and procedural changes in FDA, USEPA, and IBWA standards.

Today, a few of the water bottlers in the United States whose source water contains excessive amounts of bromide, face another challenge. Ozone can oxidize the bromide to bromate under certain conditions. Thus, the bromate concentration could exceed the newly established disinfection by-product (DBP), maximum contaminant level (MCL = 10 ug/l). Due to the relative or perceived ease of ozone treatment, and " the more ozone the better" philosophy, in many cases excessive amounts of ozone have been added to the water especially when the bottling operation is carried out without the use of ozone dosage controls necessary for the more accurate dosing of ozone. The excessive application of ozone can result in unnecessarily high bromate concentration. However, when ozone monitors and process control instrumentation are used and the ozone treatment process parameters are adjusted carefully, the bromate formation can be limited to levels below the MCL in most cases. The steps available for minimizing bromate formation are discussed in detail further below.

Brief History

Ozone treatment played a pivotal role at the beginning of the bottled water industry, and contributed to the healthy growth of the industry, which this industry has now enjoyed for many years. In the 1970's, the early years of the water bottling industry, not all the water bottlers used ozone treatment for disinfection. In addition, the water bottling process was not developed fully nor was the bottled water always sealed properly. During the handling and squeezing of the bottle, air and airborne microorganisms could enter the bottle. Thus, after days or weeks of storage, often on supermarket shelves, the potential existed for the explosive growth of microorganisms in the bottled water, which could lead to taste, odor and health problems. Several well publicized bottled water recalls took place during those days. In fact, one might say, that the newly developed ozone treatment saved the bottled water industry in the early days, in the 1970's when bottled water was not always disinfected properly, and was frequently criticized on television and in newspaper investigative reports for the rapidly deteriorating water quality during storage on the shelves of supermarkets. Shortly thereafter, under pressure from many state health organizations and the FDA, proper disinfection processes were developed for water bottling with ozone treatment as a key component. The required ozone dosages, contact times, and closure requirements for the various types of bottles and waters were determined experimentally. Ozone proved to be the magical disinfectant and oxidant that could disinfect the water, the bottling equipment, the bottle, and the sealed cap of the bottle while also oxidizing any traces of odorous materials that might be present in the water. Then the ozone decomposed to harmless oxygen and thereby disappeared without leaving an undesirable taste or odor behind.

The ozone treatment could accomplish the disinfection and the chemical oxidation of odorous materials simultaneously and enabled the water bottler to produce good quality, storage-stable water, free of objectionable by-products, taste and odor associated with the use of chlorine for disinfection/oxidation of municipal or public tap water. These much improved water quality characteristics and the claims of good-tasting, odor-free, pollution-free, healthy, storage stable water led to the rapid growth of the bottled water industry in the 1980's and 1990s with overall growth consistently approaching or exceeding double digits. (Tables I and II).

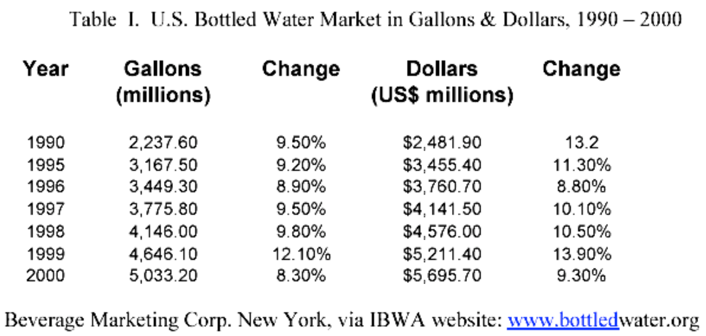


Table I. U.S. Bottled Water Market in Gallons & Dollars, 1990 – 2000

Year	Gallons (millions)	Change	Dollars (US$ millions)	Change
1990	2,237.60	9.50%	$2,481.90	13.2
1995	3,167.50	9.20%	$3,455.40	11.30%
1996	3,449.30	8.90%	$3,760.70	8.80%
1997	3,775.80	9.50%	$4,141.50	10.10%
1998	4,146.00	9.80%	$4,576.00	10.50%
1999	4,646.10	12.10%	$5,211.40	13.90%
2000	5,033.20	8.30%	$5,695.70	9.30%

Beverage Marketing Corp. New York, via IBWA website: www.bottledwater.org

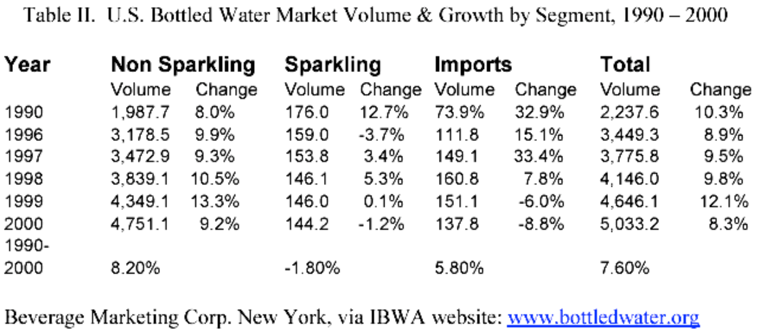


Table II. U.S. Bottled Water Market Volume & Growth by Segment, 1990 – 2000

Year	Non Sparkling		Sparkling		Imports		Total	
	Volume	Change	Volume	Change	Volume	Change	Volume	Change
1990	1,987.7	8.0%	176.0	12.7%	73.9%	32.9%	2,237.6	10.3%
1996	3,178.5	9.9%	159.0	-3.7%	111.8	15.1%	3,449.3	8.9%
1997	3,472.9	9.3%	153.8	3.4%	149.1	33.4%	3,775.8	9.5%
1998	3,839.1	10.5%	146.1	5.3%	160.8	7.8%	4,146.0	9.8%
1999	4,349.1	13.3%	146.0	0.1%	151.1	-6.0%	4,646.1	12.1%
2000	4,751.1	9.2%	144.2	-1.2%	137.8	-8.8%	5,033.2	8.3%
1990-2000	8.20%		-1.80%		5.80%		7.60%	

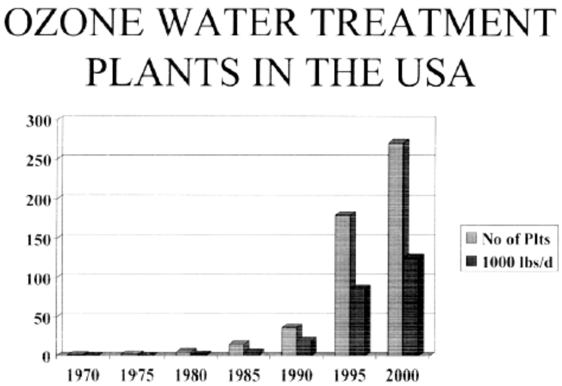
Beverage Marketing Corp. New York, via IBWA website: www.bottledwater.org

Ozone

Ozone (o3) is a gaseous material produced from oxygen in an electric discharge field (corona discharge) type ozone generator. Today, the output of the ozone generator typically contains 3-10 percent by weight (% w.t.) of ozone in the unreacted oxygen gas stream. Early ozone generators operated at 1-2% w.t. ozone concentration. The product ozone gas stream is brought into contact with the water to be treated in a device called an ozone contactor. In the ozone contactor, the ozone is dissolved in the water for the treatment, and the undissolved ozone remaining in the off-gas stream is discharged through an ozone decomposer where it is reconverted to oxygen and then released to the atmosphere at rooftop levels.

Ozone is a powerful oxidant and an exceptional chemical disinfectant. The ozone treatment process is an integral part of the drinking water treatment plant operations in more than 3,000 municipal water installations worldwide. (1). These plants supply water to the residents of many major internationally famous cities from London, Paris, Budapest, Kiev, Moscow and Singapore. In the United States alone there are nearly 400 other ozone drinking water installations including those in Dallas, Los Angeles, Milwaukee, Orlando, and Atlanta and soon in Boston and New York City. See Figure 1.

FIGURE-1



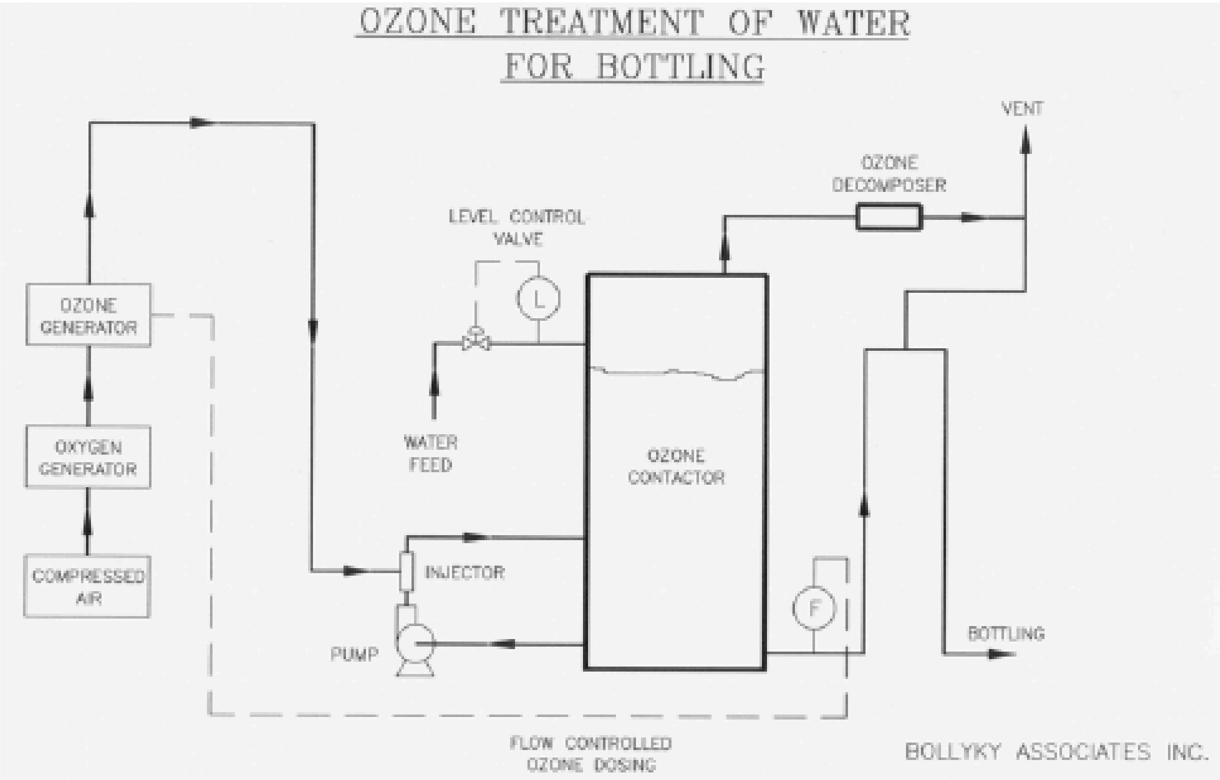
Ozone Bottled Water Treatment

The ozone is added to the water in an ozone contactor just prior to the bottling of the water. The ozone contacting system serves two primary functions (2).

Firstly, it is used for the dissolution or mass transfer of the ozone gas from the output gas mixture stream of the ozone generator into the water to be treated. A sufficient amount of ozone must be dissolved in the water to achieve the disinfection of the water and to provide dissolved ozone residual in the water necessary for the disinfection of the bottling equipment, the bottle and the cap of the sealed bottled water. In addition, dissolved ozone must be provided for the chemical oxidation of any undesirable organic or inorganic contaminants present in the water, such as, odorous materials, iron, manganese etc. The balance of the ozone remaining in the off-gas from the ozone contactor is destroyed by passing it through an ozone decomposer unit so that the off gas discharged into the atmosphere contains an ozone concentration less than 0.1 parts per million by volume (ppm v.) – or 2×10^{-4} milligrams per liter (mg/l). For the optimum treatment of the water and for minimum bromate formation, the addition of the ozone should be controlled automatically using ozone monitors and process control instrumentation. Such a treatment system should assure a sufficient dissolved ozone concentration for the treatment, but prevent an excessive amount of ozone that could facilitate by-product formation. Thus, for high quality bottled water, the addition of ozone should be carefully controlled and monitored.

Secondly, the ozone contactor is a reactor. It provides the sufficient reaction time (detention or contact time) to allow the desired disinfection and/or oxidation processes to occur in the water. The reaction time is an important parameter for all three ozone reactions encountered in water bottling: disinfection, oxidation and decomposition as discussed in detail further below. Therefore, the size or volume of the ozone contactor should be designed based on water flow, water quality, and reaction time requirements. It might be necessary for certain source waters and for some water bottling plants, that these requirements be determined experimentally by pilot plant or laboratory testing.

The other factors that influence the ozone demand and overall ozone treatment include: temperature, pH, filler design, operating procedures etc. For the best results, their effects should also be evaluated and taken into account. Process controls are an important part of a bottled water ozone treatment system. The most important of these controls is the automatic ozone dosage control. As it was pointed out above the objective of this control loop is to assure that a sufficient but not excessive amount of ozone is added to the water to obtain the required concentration of dissolved ozone in the water in order to achieve disinfection, taste, and odor control, but not produce undesirable by-products under the conditions of the water bottling operation. Some other process controls are also discussed further below. See Figures 2 and 3.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OZONE DOSING
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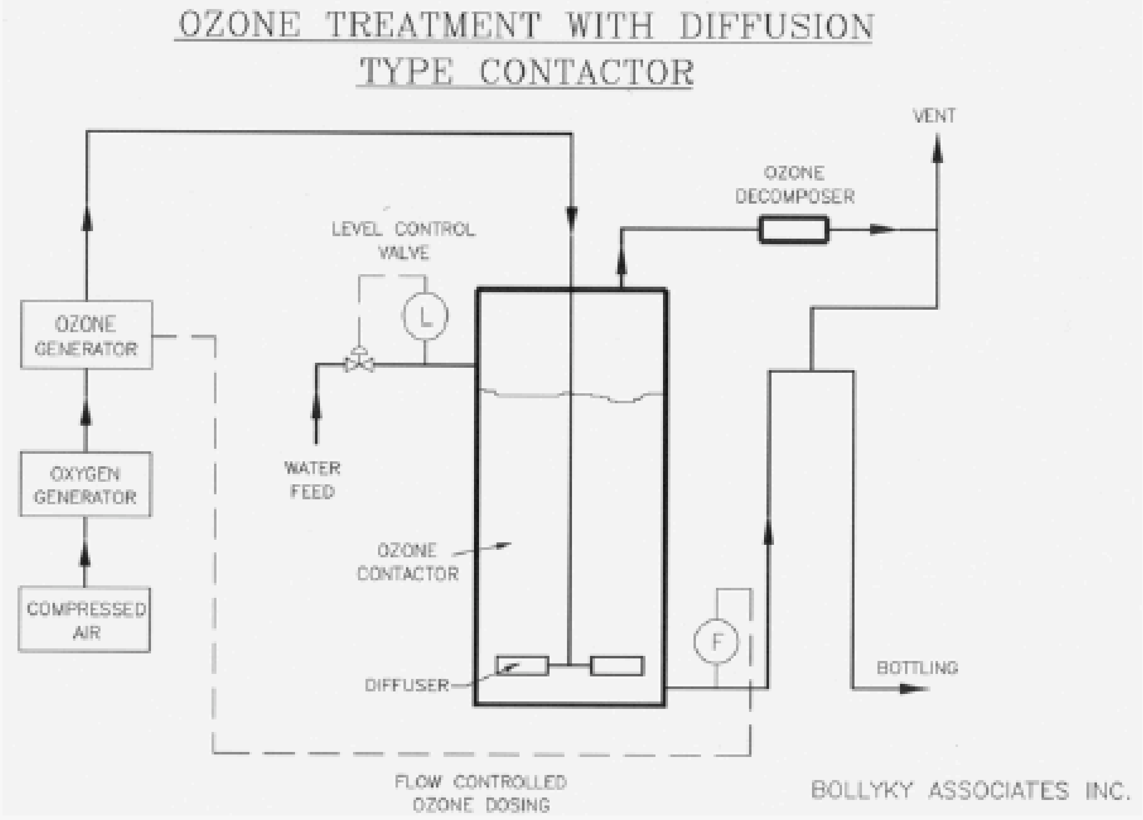
FIGURE-3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Reactions of Ozone

Once the ozone is dissolved in the water, it undergoes three simultaneous reactions. They are discussed below in detail.

1. **Disinfection:** The disinfection process treats the water against bacteria, viruses and parasites such as Giardia and Cryptosporidium. While many more details are available in the literature (1,3), it is sufficient to say here that ozone is a highly effectiv**e,** general, broad-spectrum disinfectant. It is considered to be the most effective chemical disinfectant commonly available against all the above mentioned microorganisms. See Table III. The extent of the disinfection can be defined in terms of the Ct value. The Ct value is the product of dissolved ozone concentration (in mg/l) and disinfection time (in minutes). For the disinfection effect of ozone see Tables III, IV and V.

Table III. Values of Specific Coefficients of Lethality for the Main Disinfectants (L/mg/min)

Disinfectant	Enterobacteria	Viruses	Bacterial Spores	Amoebic Cysts
O_3	500	5	2	0.5
HOCl	20	1 up	0.05	0.05
OCl^-	0.2	<0.02	<0.0005	0.0005
NH_2Cl	0.1	0.005	0.001	0.02

Source: Morris (1975)

Table IV Proposed U.S. EPA C · t Values (mg · min/L) for the Inactivation of *Giardia* Cysts with Ozone at Different Temperatures and pHs from 6 to 9

	Temperature (°C)					
Inactivation	0.5	5	10	15	20	25
0.5 log	0.48	0.32	0.23	0.16	0.12	0.08
1 log	0.97	0.63	0.48	0.32	0.24	0.16
1.5 log	1.5	0.95	0.72	0.48	0.36	0.24
2 log	1.9	1.3	0.95	0.63	0.48	0.32
2.5 log	2.4	1.6	1.2	0.79	0.60	0.40
3 log	2.9	1.9	1.4	0.95	0.72	0.46

Source. U.S. EPA (1989).

Table V C · t Values (mg · min/L) for 99 Percent Inactivation of Microorganisms with Disinfectants at 5°C

	Disinfectant			
Microorganism	Free Chlorine (pH 6 to 7)	Preformed Chloramine (pH 8 to 9)	Chloride Dioxide (pH 6 to 7)	Ozone (pH 6 to 7)
E. coli	0.034–0.05	95–180	0.4–0.75	0.02
Polio 1	1.1–2.5	770–3740	0.2–6.7	0.1–0.2
Rotavirus	0.01–0.05	3810–6480	0.2–2.1	0.006–0.06
Phage f2	0.08–0.18	–	–	–
G. lamblia cysts	47–>150	–	–	0.5–0.6
G. muris cysts	30–630	1400	7.2–18.5	1.8–2.0

Source: Hoff (1987).

The relative disinfection effects, that is the values of the Specific Coefficient of Lethality of four disinfectants are shown in Table III on four microorganisms. The data in Table III indicate that ozone is 5-25 times more effective against bacteria, viruses, spores and cysts than the various chlorine type disinfectants, such as, hypochlorons acid, hypochlorite, or chloramine.

The Ct values proposed by the EPA for the disinfection of Giardia are displayed in Table IV. These data indicate the recommended Ct. values for the various degrees of inactivation under six temperature conditions.

The required Ct. values for 99% inactivation of six different microorganisms are summarized in Table V. The data in Table V provide the recommended Ct values for the selected group of four different disinfectants including ozone. In general, the disinfection by ozone requires significantly lower Ct. values than by the other three disinfectants covered. They are: chlorine, chloramines or chlorine dioxide. This fact is due to the substantially more powerful disinfection effect of ozone.

2. **Chemical Oxidation:** Ozone is a powerful oxidizing agent. It is very effective in water treatment against essentially all taste and odor causing organic materials and oxidizable inorganics like iron, manganese and sulfide ion. Again, these reactions have been studied in great detail and are reported on in the literature. (4). The required ozone concentrations and reaction times are dependent on the type and concentration of the oxidizable pollutants present in the water and on the water quality. They should be determined experimentally in a treatability study.

3. **Decomposition:** ozone is an unstable compound under room temperature and near room temperature conditions and decomposes rapidly to oxygen. The decomposition rate is influenced primarily by water temperature and pH. The half-life of ozone at 20 $_{o}$C and pH 7.0 is typically 24 minutes in potable water. See Figure 4. In most cases there is ample time available for the ozone decomposition to take place before the bottled water is delivered to the consumer. Typically, 5-10 hrs should be sufficient. More specifically, the decomposition rate is dependant on pH, temperature and the type and concentration of inorganic salts present in the water.

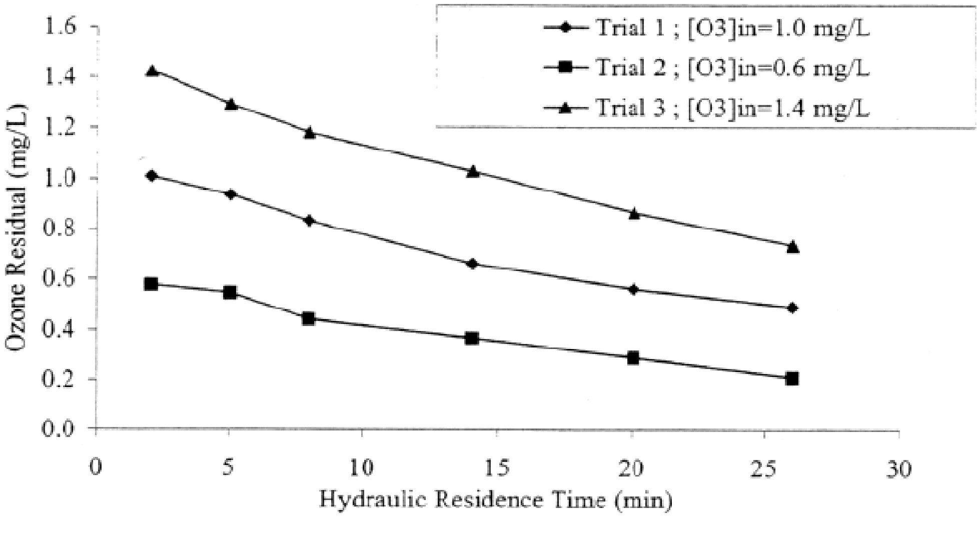


Figure 4 Ozone decay profiles at pilot-scale at Chertsey WTP (UK)
[O3]in: ozone residual at the inlet of the contactor (or outlet of the gas-liquid separator)
Chandrakanth, 2001

Regulatory Oversight

The Food and Drug Administration (FDA) has the regulatory and oversight responsibility over the bottled water in the USA. Title 21 of the Code of Federal Regulations (CFR) under Section of Part 165 outlines the required physical, chemical, radiological and microbiological parameters for bottled water (5). The FDA automatically adopts the applicable improvements and changes in drinking water quality requirements set by the EPA. The IBWA standards are based on FDA and EPA regulations.

The FDA has granted GRAS status for ozone disinfection in November 1982. According to the FDA: “a substance added to the food (and water is a food) may be marketed without a regulation if it is one which is considered as Generally Recognized As Safe (GRAS) by experts qualified by scientific training and experience to evaluate safety”. Thus, the FDA classified ozone as Generally Recognized As Safe” (GRAS) for use as a disinfectant for bottled water up to a residual dissolved ozone concentration of 0.4 mg/l. Furthermore, the FDA has recognized ozone treatment to be a Good Manufacturing Practice (GMP) for the bottled water industry. This regulation appears in Code 21 of Federal Regulations, Section 129.80 d.4 and first appeared in the Federal Register 11566, 12 March 1975. The minimum ozone treatment for GMP is “0.1 part per million (0.1 mg/l) ozone in water solution in an enclosed systems for at least 5 minutes.

The FDA has jurisdiction also about the allowed level of ozone concentration in the air or atmosphere of residential areas. The allowed maximum ozone concentration is 0.05 ppm by volume.

The Department of Labor and its agency, the Occupational Safety and Health Administration (OSHA) has the regulatory authority over the allowed maximum ozone concentration levels in the atmosphere of work places, such as, water bottling plants. The maximum ozone concentration permitted in those places is 0.1 ppm by volume. The recommended practice is the installation of an ambient air ozone safety system consisting of an ozone monitor coupled to the on/off control of the ozone generator and of the room’s exhaust air system. When the ozone concentration exceeds the allowed preset level, the safety system shuts off the ozone generator and activates the air exhaust system. In order to meet the above OSHA regulatory requirement, the ozone treatment system Should be a closed system with a vent for the off-gas to outdoors at roof top level through an ozone decomposer. The filler should be hooded or enclosed with an exhaust to the outdoors.

The International Bottled Water Association (IBWA), although it is not a regulatory agency, it offers a manual to its corporate members called the “IBWA Plant Technical Reference Manual”. (6). This manual covers among others guidelines, standards and OSHA recording and reporting requirements.

Disinfection By-Products

The water disinfection process should be applied downstream of all water treatment and water conditioning of the source water just prior to the bottling step of the bottled water production (6). The disinfection process is limited to smaller microorganisms such as bacteria and viruses. The disinfection of larger parasites, such as, Cryptosporidium is not required by the FDA since 75% of the water bottlers use ground water as their source water which is not expected to contain Cryptosporidium and the other 25% use the public water supply that is already treated for it. The ozone disinfection of bottled water is expected to treat the smaller microorganisms with dissolved ozone concentrations in the range of 0.1 – 0.4 mg/l. The required Ct values for six microorganisms are shown in Tables IV and V. During an estimated 20 minutes overall contact time, the dissolved ozone concentration of 0.1 mg/l and 0.4 mg/l will provide Ct values of 2.0 and 8.0

respectively. A possible complication is that the dissolved ozone may oxidize the bromide ion present in the source water to bromate during that time. The FDA has set a maximum concentration limit, MCL = 10 ug/l, for this disinfection by-product as of January 2001 (Federal Register, July 5, 2001). This limit is the same as the one set by the EPA for public drinking water. Thus, the 25% of the water bottlers using the public water for their source water will automatically meet this DBP (disinfection by-product) requirement.

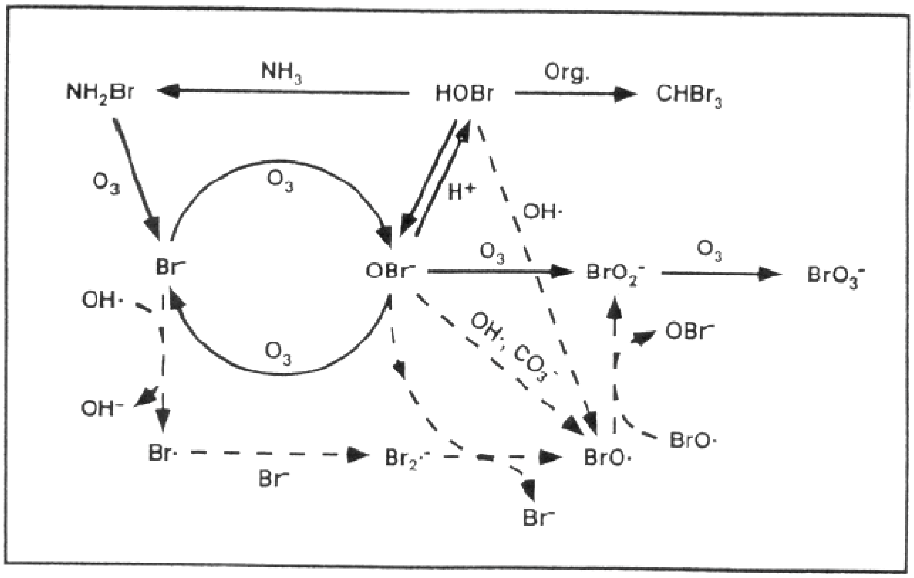
The amount of bromate by-product that can be produced during ozone disinfection is dependent on the concentration of the bromide in the source water. As a good indication, we might consider that essentially all the US water treatment plants using ozone treatment that have 60 ug/l bromide or less in their raw water routinely meet the bromate MCL with ease. Furthermore, there are steps that can be taken for the control of bromate formation during the ozone disinfection process. They are outlined below. An important aspect of the FDA ruling on July 5, 2001 is that the FDA will not require the bottled water already on the market to be recalled before the two year shelf life expires. This ruling allows time for the evaluation and modification of the ozone disinfection process should it be necessary to do so.

Bromate Control

The ozone disinfection of source water containing a certain concentration of bromide will be dependent on the magnitude of the bromide concentration and on the water quality of that water such as pH, alkalinity, the concentration and type of organic and inorganic compounds present in the water and on the water temperature.

The formation of the bromate consists of a complex sequence of multiple reactions. It has been studied in great detail (7) and can be summarized as shown in Figure 5 and Table VI. As Figure 5 indicates, the bromide is oxidized by ozone first to hydrobromous acid. The hydrobromous acid dissociates into hydrobromite ion to an extent depending on the pH of the water. The hydrobromite then can be oxidized further either by ozone or hydroxyl radical. The final oxidation product is bromate. The individual reactions and their rates are shown in Table VI.

Fig. 5 Scheme of reactions of ozone and OH radicals in bromide-containing waters



(from: U. von Gunten and J. Hoigné,

Table VI Reactions and rate constants involved in bromate formation by molecular ozone pathway

	Reaction	k or pKa (20°C)	Reference
1	$O_3 + Br^- \rightarrow O_2 + BrO^-$	$160\ M^{-1}.s^{-1}$	a
2	$O_3 + BrO^- \rightarrow 2\ O_2 + Br^-$	$330\ M^{-1}.s^{-1}$	a
3	$O_3 + BrO^- \rightarrow BrO_2^- + O_2$	$100\ M^{-1}.s^{-1}$	a
4	$O_3 + HOBr \rightarrow BrO_2^- + O_2 + H^+$	$\leq 0.013\ M^{-1}.s^{-1}$	a
5	$BrO_2^- + O_3 \rightarrow BrO_3^- + O_2$	$>10^5\ M^{-1}.s^{-1}$	a
6	$HOBr + NH_3 \rightarrow NH_2Br + H_2O$	$8\ 10^7 M^{-1}s^{-1}$	b
7	$HOBr + NH_2Br \rightarrow NHBr_2 + H_2O$	$7\ 10^5\ M^{-1}s^{-1}$	c
8	$HOBr + NHBr_2 \rightarrow NBr_3 + H_2O$	$2.5\ 10^4\ M^{-1}s^{-1}$	c
9	$3\ O_3 + NH_2Br \rightarrow 2\ H^+ + NO_3^- + Br^- + 3\ O_2$	$40\ M^{-1}s^{-1}$	b
10	$O_3 + NHBr_2 \rightarrow ?$	$10\ M^{-1}s^{-1}$	b
11	$O_3 + NBr_3 \rightarrow ?$	?	
12	$HOBr \leftrightarrow H^+ + BrO^-$	8.7	
13	$NH_4^+ \leftrightarrow H^+ + NH_3$	9.3	

a : Haag and Hoigné (1983) b : Haag *et al.*, 1984 c : Haag and Lietzke (1980)

The formation of bromate can be lowered by the addition of ammonia, organic and inorganic free radical inhibitors, lowering pH and temperature as Figure 6 and 7 indicate. The lower pH reduces the amount of hypobromite present by shifting the equilibrium to hypobromous acid, and, at the same time, it lowers the hydroxyl radical formation from ozone. As Figure 6 indicates, the lower pH results in lower bromate formation. On the other hand, the greater exposure to ozone produces more bromate as Figure 7 shows. Furthermore, the higher water temperature also favors bromate formation.

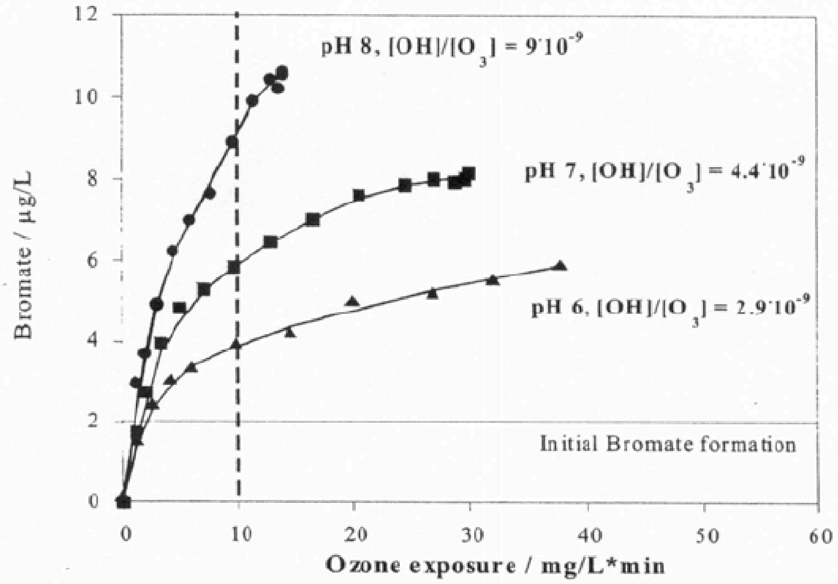


Fig.6 Bromate formation as a function of the ozone exposure for the pH range 6 – 8.
experiments were performed in river Seine water spiked to a bromide concentration of
µg/L. U. V. Gunten,2001

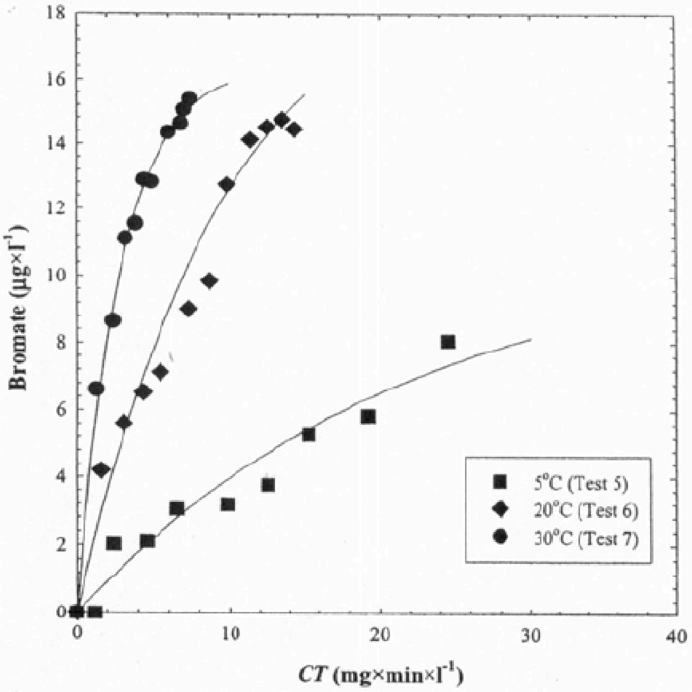


Fig.7 Bromate formation as a function of temperature in Lake Zürich water at pH 7. $[Br^-]_o$ = 33 µg/L. Adapted from Driedger et al. (2001).

Many studies have been carried out on natural raw water sources to find the most effective way of controlling the formation of bromate. (8) The consensus of these studies is that in general the following steps can be taken:

1. The most effective step by far is the control of ozone dosage to a minimum dissolved ozone concentration required for the disinfection process during the entire water bottling operation. The properly designed bottling system should provide for the needed ozone concentration and ozone contact time (Ct) in the ozone contactor using ozone monitors and automatic controls. The design considerations should also take into account the ozone concentration and reaction time occurring in the bottle. The step of accurate, automatic ozone dosage control is a key step. It does not require the addition of any chemicals and still can be the most effective step for bromate control. It is highly recommended and should be considered and tried first.
2. The second most effective step is the lowering of the pH. The formation of bromate can be reduced significantly when the addition of a suitable acid lowers the pH for example from pH = 7.5 to pH = 6.0. However, the adjustment of the pH requires a carefully designed and operated pH control system consisting of a metering pump, mixer, tanks, and pH meter and a control system. The system should be designed properly and be operated and monitored with care. The small change in the pH does not affect the disinfection significantly but can reduce bromate formation substantially.
3. The third most effective step is the lowering of the water temperature. Under the lower water temperature condition, less bromate is produced, but the ozone disinfection process is not affected significantly. However, the drawback of this

step is the significant energy input that might be needed for the cooling of the water. The refrigeration equipment is readily available commercially.

4. The fourth bromate lowering step is the addition of a small dose of ammonia. As indicated in Figure 5, the ammonia reacts with the hypobromons acid and renders smaller amounts of the hypobromate available for oxidation to bromate. For the treatment of most source waters with significant bromide concentrations the above recommended optional Step 1, the automatic control of ozone dosing should be sufficient to maintain the bromate formation below the MCL = 10 ug/l level. Should there be further steps required, we recommend a careful experimental evaluation of the treatment options available in a pilot plant study on site or in a more economical laboratory bench scale study on water samples shipped overnight on ice to a qualified laboratory. Contact the International Ozone Association for a list of Laboratories and Consultants.

Conclusion

The ozone treatment is essential for water bottling because it plays a multiple, beneficial role in the production of bottled water and can when, properly done, assure a good quality, storage stable product. Ozone is the most powerful, chemical disinfectant available. It disinfects them all: the water, the bottle, the bottling equipment, the sealed cap of the bottle and any air borne micro organisms in the air space above the water. Furthermore, it improves, taste, eliminates odor and oxidizes undesirable organic and inorganic materials that might be present in the source water. Then, once ozone has done its job, it decomposes to harmless oxygen. Should you wish for an ideal disinfectant for bottled water, you could not dream of a better one than ozone.

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