TREATMENT OF PROCESS WASTEWATERS FOR RECYCLING USING AN ADVANCED OXIDATION PROCESS

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Abstract. UV-enhanced ozonation experiments on unscreened overflow chiller-water samples from a commercial poultry-processing plant successfully compared the oxidative and bactericidal effects of four treatments (*viz.*, O_2/O_3, O_2/UV, O_2/O_3/UV, and O_2 as the control). At least a 60% reduction of total microorganisms including similar reductions in coliforms and *E. coli* as well as the maintenance of light transmission at a value no less than 80% that of fresh water were obtained. This satisfies USDA-FSIS regulations that permit 90% recirculation of 0.5 gal (1.9 L) overflow per carcass required. Also an additional reduction (>0.8 Log CFU/mL) in aerobic plate count (APC) was documented for ozone acting in concert with UV photons as compared with the sum of the effects of O_3 and UV acting separately in series. Economic analysis of operational and maintenance costs for the UV-enhanced ozonation system recommended from this work estimates annual savings of ~$244,000 in a 1/4-million bird per day plant achieving water savings of 426,000 L/day (113,000 gal/day).

INTRODUCTION

This paper reports an advanced oxidation process (AOP) of ultraviolet-photon enhanced ozonation investigated for improving microbiological safety, turbidity, and water-use efficiency of overflow poultry chiller water allowing its reconditioning for reuse.

The poultry-processing industry in Georgia is critically challenged by competitive economic pressures, food safety and quality issues, and environmental constraints — major ones being excessive use and pollution of water. Conservative estimates of yearly water use by sampled American poultry processing plants ranged over 40-500 million gallons per plant; mostly 125-175 million gallons (Simon, 1985). Due to the detrimental mixing of pathogenic microbes occurring in bath-type carcass chilling systems, a portion of the water has to be continuously removed (0.5 gal overflow per carcass).

Poultry processors need to decrease water consumption by implementing conservation practices. USDA's Food Safety and Inspection Service (FSIS) regulations provide incentives for chiller-water reconditioning by way of an amendment to the Federal Poultry Products Inspection Act (USDA, 1997). This ruling permits water reconditioning if there is at least a 60% reduction of total microorganisms including similar reductions (*i.e.*, within ± 10%) of coliforms, *Escherichia coli* (*E. coli*), and *Salmonella spp.*, as well as the maintenance of light transmission (at 500 nm) at a value no less than 60% that of fresh water.

BACKGROUND AND RELATED WORK

Many water-reconditioning methods have been developed. Filtration methods with chlorination have been widely used (Chang *et al.*, 1989). Chlorination has the adverse effect of producing carcinogenic chlorinated hydrocarbon by-products (*e.g.*, trihalomethanes) in water. Filters provide very short filtration cycles (*e.g.*, less than 20 min) that require additional mechanization in order to remove material buildup. Li *et al.* (1994) achieved significant reductions in *Salmonella* typhimurium by applying pulsed electric current through poultry chiller water. In order for this type of system to work at low voltages for worker safety, salt needs to be added to improve conductivity. Increased salt concentrations could detrimentally affect poultry carcasses. Other chemicals have been utilized but have degraded broiler carcass appearance (Li *et al.*, 1995). With the exception of *Salmonella*, Izat *et al.* (1990) found ozone to meet current USDA guidelines for pathogen reduction. They proposed combining ozone with another treatment such as UV radiation or hydrogen peroxide (H_2O_2), could produce an effective water treatment system.

UV radiation has been successfully used for wastewater treatment (Prengle *et al.*, 1975). A typical and effective application for wastewater reuse is that of filtered activated sludge effluent. Braunstein *et al.* (1996) found that a UV disinfection system, using filtered secondary effluent under field conditions at a wastewater treatment facility, was technically feasible even with the most stringent wastewater reclamation requirements of California for spray irrigation of food crops and nonrestricted recreational impoundments. Other studies have shown that the microbiological quality of broiler carcasses has improved by reduction of *Salmonella* surface contamination without negatively affecting carcass color or increasing rancidity of the meat when irradiated with UV energy. This process has several advantages: simple operation, no harmful residuals, operator safety, no intermediate chemical compounds, low capital cost, and lenient regulatory constraints (EPRI, 1993 and Hill, 1997). One disadvantage is that turbidity in water (such as chill water) can reduce its efficacy by UV attenuation.

Ozone provides several advantages over conventional chlorine water treatment. Lin and Yeh (1993) mentioned how much safer it is to use than chlorine (ozone being made on site at low pressures and easy to stop its production). Ozone is effective for organic and inorganic oxidation, color, odor, and suspended-material removal, disinfection, deodorization, bleaching, abatement of environmental pollutants, and algal growth control. In early 1991 USDA granted the first application and approval of a recycling system to treat overflow chiller water using ozone after all the data collected by Waldroup *et al.*(1993) provided final documentation criteria to obtain approval. Increase in % light transmission and reductions in microorganisms for the turkey processing plant met USDA recycling specifications and allowed maximum recycling rates (90% of chiller water). Much prior internationally-based work has confirmed ozone's ability, even at less than 1 mg/L (*i.e.*, 1 ppm) dissolved concentrations, to deactivate numerous water-borne pathogenic microorganisms, including those commonly found in poultry processing waters, and to oxidize suspended organic solids. The future for ozone use in the food processing industry as a food disinfectant or sanitizer appears even more promising after an expert panel convened by the Electric Power Research Institute (EPRI) prompted the Food and Drug Administration (FDA) to declare ozone to be generally recognized as safe (GRAS) (Graham, 1997). Hence ozonation offers much potential for water conservation as well as enhanced waste treatment of processing waters.

EXPERIMENTAL DESIGN

In this paper the proposed treatment method is an AOP of UV-enhanced ozonation (O_3/UV) for improving the microbiological safety, turbidity, and water-use efficiency of overflow poultry chill water allowing its reconditioning for reuse. Ozone alone not only has 1.52-times the oxidation potential of chlorine, but as it is excited by UV photons, it decomposes into free radicals, hydroxyl (OH·) and oxygen [$O(^1\Delta)$, $O(^3P)$, $O(^1D)$], each having a higher oxidation potential than either ozone or chlorine (Lin and Yeh, 1993). With ozone's peak radiant absorption between 253-255 nm wavelength and short-wave UV lamps providing a strong output band at 253.7 nm, reactions can be enhanced 10^2-10^4 fold due to the direct formation of additional radicals by UV photons as well as an increase in ozone reaction (Prengle *et al.*, 1975). UV irradiation also generates more free radicals than ozone from neutral and refractory molecules thus making the overall reaction go much faster.

The experimental plan had two phases. The first phase involved tests utilizing broiler overflow chiller water samples to understand the enhanced effect of the ozone-UV process under replicable conditions. The second phase involved determining the effectiveness of the O_3/UV process in eliminating *Salmonella* from chiller water.

METHODS

Chiller Wastewater Trial

Utilizing the knowledge obtained from previous work (Diaz and Law, 1997), this phase of experiments treated overflow chiller water from a number of sampling days. Overflow chiller water samples were obtained from a nearby broiler processing plant during the period of March through November 1997. Sampling done prior to the experimental trial at different daily periods and stages of the processing cycle indicated wide variability in the water quality parameters. Peak microbial and organic levels occurred after the last carcass of the second shift entered the pre-chiller prior to the sanitation shift (*i.e.*, when the chillers were emptied and cleaned). It is at this sampling time that the real-life efficacy and efficiency of the AOP process in treating poultry overflow chiller water was evaluated.

The experiments were performed for random treatment combinations of three ozone and UV levels, examined at contact times of 4, 8, and 16 min. The applied ozone dose levels were 0, 2.55±0.21, and 5.11±0.42 mg/L·min; the UV power levels were 0, 295 and 428 mW/L. A 10 L batch of overflow chiller water was transported to the laboratory and maintained at 4°C. Before and after treatment the batch was sampled to determine water quality parameters: total suspended solids (TSS), percent light transmission (%LT) at 500 nm, chemical oxygen demand (COD), *E. coli*, total coliforms (TC), and aerobic plate count (APC).

The stock sample was used to obtain 250 mL sample volumes per reactor which comprised a single experimental sample. Three grouped reactors comprised a single experimental run or block. Nine of these blocks made up a single experimental replication. A factorial experimental design allowed testing for interactions with at least three replications comprising a cell or block. Three factors at different levels each were established prior to commencing treatment and evaluated following it: ozone dosage, UV radiation intensity, and contact time. Responses measured following treatment were the water quality parameters already mentioned.

This phase showed the effectiveness and kinetics of the system utilizing process water. It determined the ozone and UV combination that, within the least amount of time, disinfected the chiller water to meet current USDA regulations. Also, this phase determined how changes in ozone concentrations and UV intensities affected the microbial concentration and light transmission of poultry wastewater.

Salmonella typhimurium Trial

According to Campbell *et al.* (1982) and Lillard (1978), *Salmonella spp.* numbers in pre-chiller overflow water are usually very low or non-existent (*i.e.*, less than Log 0.4-1.0 CFU/mL). It was necessary to numerically quantify the reductions in *Salmonella* before and after treatment without the use of expensive antigen detection kits that screen foods and feeds for the presence of the *Salmonella* species and only provide culture confirmation (*i.e.*, presumptive positive). An alternative method was to use a modified strain of Nalidixic Acid (NAL) resistant *Salmonella* (easily selected upon growth on a selective medium) to be used for inoculating the wastewater and then easily quantified.

The strain was inoculated from two beads, kept in a cryoprotective medium at -80°C, into a tube containing 9 mL of tryptic soya broth (TSB) containing ~200 ppm of NAL and then incubated at 35°C for 24 h. From this culture 1 mL was transferred into another TSB+NAL tube and incubated at 35°C for 24 h; *the S. typhimurium* were then acclimated to cold temperatures at 4-8°C for 24 h. Before the experiment, 10 mL of this suspension was diluted 1:1000 with 10 L of previously sampled broiler overflow chiller wastewater (obtained earlier from the plant and stored at 4°C) to obtain an initial plate count of Log 5.5 CFU/mL. The inoculated wastewater was kept at 4°C to maintain constant *S. typhimurium* numbers throughout the experiment. The 10 L stock sample was used to obtain 250 mL sample volumes per reactor. Each 250 mL volume of pre-chiller overflow wastewater comprised a single experimental sample. Six grouped reactors comprised a single experimental run or block. Two of these blocks made up a single experimental replication. This phase utilized the optimal settings of UV and ozone dosages obtained in the previous phase (*i.e.*, 5.11 mg/L·min and 295 mW/L, respectively). *S. typhimurium* counts were recorded as the number of CFUs per plate. Counts were statistically analyzed by GLM procedure of SAS (SAS Institute, 1996) after a log transformation; this was necessary in order to bring the distribution of the data closer to normality since microbiological data in its raw form does not produce the normal distribution needed for meaningful statistical analysis. A minimum of three replicates were needed to obtain a balanced 3×2×2 factorial experiment in blocks of size six as described by Winer (1962). Comparisons between O_3 vs. UV, O_3 vs. O_3/UV, and UV vs. O_3/UV were performed.

This phase determined if ozone with UV is effective for reducing or eliminating *Salmonella* in poultry chiller overflow water since USDA (1997) recycling standards requires the reduction of least 60% of total microorganisms including *Salmonella spp.* in poultry overflow chiller water.

Table 1. Effect of different treatment combinations of O_3 and UV on several water quality parameters in broiler overflow chiller wastewater after 16 minutes of treatment duration.[a]

Treatment	TSS (mg/L)	%LT	COD (mg/L)	*E. coli* (Log CFU/mL)	TC (Log CFU/mL)	APC (Log CFU/mL)
O_2	249	55.7	1498	2.3	2.6	4.1
O_2/O_3	122	74.5	1066	0.7	0.9	3.4
O_2/UV	248	55.2	1494	0.7	0.8	2.7
O_2/O_3/UV	123	80.4	1068	0.7	0.7	1.1

[a] Ozone dosage of 5.11 mg/L·min and UV dosage of 295 mW/L. Initial chiller water concentrations: TSS = 361 mg/L; % LT = 50.8; COD 1731 mg/L; E. coli = Log 2.3 CFU/mL; TC = Log 2.6 CFU/mL; APC = Log 4.1 CFU/mL

Table 2. Effect of the UV-enhanced ozonation on several water quality parameters in broiler overflow chiller wastewater as functions of treatment duration.[a]

Time (min)	TSS (mg/L)	%LT	COD (mg/L)	*E. coli* (Log CFU/mL)	TC (Log CFU/mL)	APC (Log CFU/mL)
0	361	50.8	1731	2.3	2.6	4.1
4	194	55.2	1285	0.7	0.8	2.5
8	167	62.0	1170	0.7	0.7	1.6
16	123	80.4	1068	0.7	0.7	1.1

[a] Ozone dosage of 5.11 mg/L·min and UV dosage of 295 mW/L.

Ozonation apparatus

A laboratory ozone generator capable of producing 0.45 kg (1 lb) per day of ozone was used to generate the ozone/oxygen gas mixture (Diaz and Law, 1997). A feedgas flow of ~9.4 L/min (20 SCFH) was provided from compressed, filtered, and dried oxygen at 83 kPa (12 psig) within the generator's electric-discharge cell. The generator was set to produce 0.500 wt% gaseous ozone concentration from oxygen feedgas as monitored with a high-concentration UV-absorption type photometric ozone monitor. As the gas diffused through a 60 mm diameter, coarse (40-60 μm nominal pore size) fritted disc at the bottom of the reactor vessel, the resulting ascending bubbles came into contact with the sample liquid. Inside a quartz glass central tube within the vessel, a quartz pencil-type irradiator lamp provided known UV radiation levels (2.55 and 5.11 mg/L·min). Two different nominal intensities were provided (4500 and 9000 $\mu W/cm^2$ at 19 mm test distance) by these low pressure, mercury vapor, cold cathode, double bore, gas discharge lamps. The 9,000 $\mu W/cm^2$ lamps (55 mm lighted length) were covered with a double layer of copper tape to obtain different intensities (*i.e.*, 25 and 18 mm lighted lengths). The UV pencil lamps provide a distinct emission peak at 253.7 nm, which is close to ozone's peak absorption at 255.3 nm. The lamp intensities were monitored before each experimental setup with a UV digital radiometer.

CONCLUSIONS

Chiller Wastewater Trial

A synergistic effect (*i.e.*, >0.8 Log CFU/mL additional reduction) in APC was obtained for ozone acting in concert with UV photons as compared with the sum of the effects of O_3 and UV acting separately (Table 1). As expected, no reductions in APC, *E. coli*, and TC resulted from only the background oxygen flow. UV dosages of 178 and 247 mW/L very effectively eliminated bacterial counts (>90% for *E. coli*, TC, and APC) for all treatment durations tested. Even though, at the dosages tested, ozone alone was not as effective as UV in reducing the microbial load, both dosages did provide >90% reduction in *E. coli* and TC and >80% reduction in APC after 16 min of treatment. During the same treatment time with an ozone dosage of 5.11 mg/L·min, an apparent synergistic effect between O_3 and UV dosage of 295 mW/L was obtained in APC reduction (Table 2). Roche *et al.* (1994) reported that in states of continuous operation the transfer of ozone could be total, thus indicating that all the ozone is being consumed by the oxidizable species in the water (organic matter, impurities, H_2O_2, OH-, etc.). The above results explain the reduced bactericidal effect of ozone in the presence of soluble organic materials as reported by Yang and Chen (1979). As more ozone is being utilized and self-decomposed in the presence of organic matter, less becomes available to impact the microbiology of the water. The most effective settings of ozone and UV dosages for this test phase were 5.11 mg/L·min and 295 mW/L, respectively. These dosages were thus used in the third and last phase for the reduction of *Salmonella typhimurium*.

A decrease in TSS and COD (361 to 249 mg/L and 1731 to 1498 mg/L, respectively) and a small increase in %LT were documented (50.8 to 55.7) after 16 minutes of treatment with only background oxygen flow. Similar results were obtained in TSS, %LT, and COD with UV dosages of 178 and 247 mW/L for all treatment durations tested. Both ozone dosages were very effective in reducing TSS (<132 mg/L) and COD (<1200 mg/L), and increasing %LT (>70%) after 16 min of treatment. For recycling, USDA regulations require at least 60% light transmission (500 nm) of treated water as compared with fresh plant water.

***Salmonella typhimurium* Trial**

The antimicrobial effects of the best settings of UV and ozone dosages, obtained in the previous phase (*i.e.*, 5.11 mg/L·min and 295 mW/L, respectively), against *Salmonella* are plotted in Fig. 1 as functions of treatment duration. Reductions of >0.9 Log (*i.e.*, 87.4%) after 16 min of ozonation were achieved using 5.11 mg/L·min; in comparison the UV intensities utilized were more effective by providing 2.6 Log (>99.9%) reductions in *Salmonella* with 295 mW/L dose after 16 min.

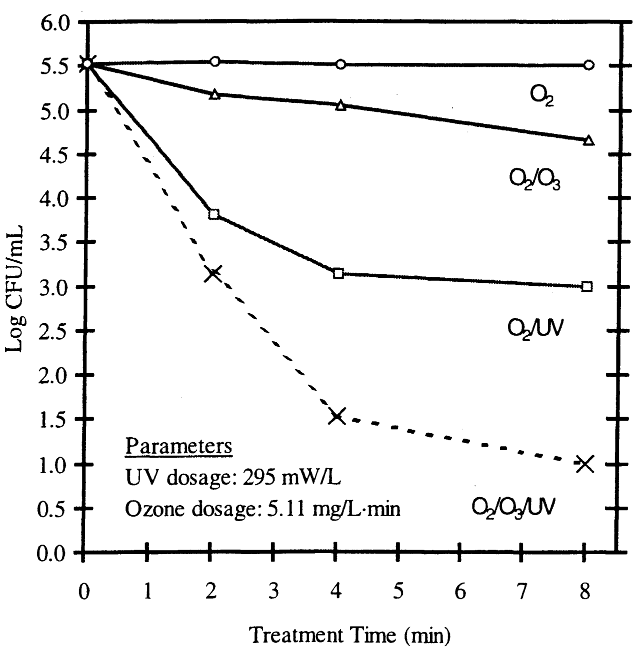

Figure 1. Bactericidal effect of three different treatments on NAL acid resistant *Salmonella typhimurium* inoculated broiler overflow chiller wastewater as functions of treatment duration.

At 8 to 16 min treatment times a synergistic bactericidal effect was documented between O_3 and UV acting simultaneously. This accounts for an additional reduction in *Salmonella* (>4.5 Log). As confirmed in Fig. 1, no reduction resulted from only the background oxygen flow and magnetic stirring. Statistical analysis revealed significant differences ($P<0.01$) for all main effects as well as their interactions. Of all the interaction effects, Ozone×UV was greatest, again suggesting that a synergistic effect between ozone and UV irradiation occurred.

DISCUSSION

This research investigated an AOP of UV-enhanced ozonation as a proposed treatment method for improving the microbiological safety, turbidity, and water-use efficiency of overflow poultry chiller water allowing its reconditioning for reuse. Gas washing reactors of 350 mL simultaneously provide independently selectable input levels of ozone and UV to facilitate such experiments. UV-enhanced ozonation treatments of unscreened poultry overflow chiller-water samples from a local processing plant have successfully compared the oxidative and bactericidal effects of four different treatments (*viz.*, O_2/O_3, O_2 /UV, O_2 /O_3 /UV, and O_2 as the control). At least a 60% reduction of total microorganisms including similar reductions in coliforms and *E. coli* as well as the maintenance of light transmission at a value no less than 80% that of fresh water were obtained. An additional reduction (>0.8 Log CFU/mL) in APC has now been documented for ozone acting in concert with UV photons as compared with the sum of the effects of O_3 and UV acting separately. Even though ozone alone, at the dosages tested, was not as effective as UV in reducing the microbial load, both ozone dosages did provide >90% reduction in *E. coli* and TC and >80% reduction in APC after 16 min of treatment. Both ozone dosages were very effective in reducing TSS (<132 mg/L) and COD (<1200 mg/L), and increasing %LT (>70%) after 16 min of treatment. During the same treatment time with an ozone dosage of 5.10 mg/L·min, an apparent synergistic effect between O_3 and UV dosage of 178 mW/L was obtained in APC reduction. These results indicate a 90% closed-loop chill-water recirculation could be obtained according to USDA-FSIS guidelines.

The second phase reduced NAL resistant *Salmonella typhimurium* inoculated in poultry overflow chiller water. Reductions of >0.9 Log (*i.e.*, 87.4%) and 2.6 Log (99.9%) after 16 min of ozonation were achieved using 5.11 mg/L·min ozone and 295 mW/L UV, respectively. A synergistic bactericidal effect was documented between O_3 and UV acting simultaneously at 8 to 16 min.

Economic analysis for operational and maintenance costs for a UV-enhanced ozonation system using of the best settings of ozone and UV dosages from the Chiller Wastewater test phase estimated savings of ~$244,000/year in a 1/4-million bird per day plant at 90% closed-loop chill-water recirculation. This included refrigeration and water/sewer costs for water savings of 426,000 L/day (*i.e.*, 113,000 gal/day). These calculations do not take into account indirect benefits achieved by lowering municipal water demand by reallocating some of it for the general public as well as reducing pollutant release into our environment. Additional economical benefits for the plant could come from flocculant rendering (Waldroup *et al.*, 1993). Savings should maintain bird price by lowering production costs without sacrificing quality.

These results portend well regarding the likely success and economic feasibility of the AOP for use not only in the poultry processing industry but others where safe, effective, and environmentally conscious treatment of wastewater is needed.

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