

**Quarterly Progress Report for
USEPA Grant S-82874601-1**

**Evaluate Pilot and Full-Scale Treatment Processes
to Remove TBT from Industrial Wastewater**

Submitted to:

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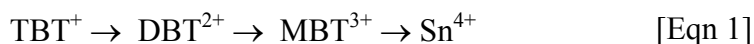
August 15, 2002

Summary of Recent Study Efforts

Research efforts in the most recent quarter have been directed toward the continued evaluation of TBT removal by ozone and ultraviolet (UV) irradiation, evaluation of GAC removal of copper and zinc using a continuous-flow column, and a laboratory GAC column study to characterize the TBT breakthrough curve and assess the TBT adsorption capacity. Experiments with UV and ozone were conducted to determine the effects of temperature on TBT removal and through the addition of hydrogen peroxide, what improvements in TBT removal could be produced. Study efforts with UV and ozone have been aided by collaborative efforts under a Sea Grant award to principal investigator Dr. Michael Unger, Virginia Institute of Marine Sciences, and G. Schafran (Co-PI) to conduct mechanistic evaluations of TBT removal.

TBT Removal by Oxidation with Ozone

All experiments reported here were conducted utilizing the laboratory, pilot-ozone treatment system housed at the environmental engineering laboratory at Old Dominion University (Figure 1). The system consists of a one-pound per day ozone generator¹ fed by pure oxygen gas that produces a high concentration ozone gas stream (up to 10% O₃). The feed gas concentration is monitored by a gas-phase ozone monitor² and aqueous phase concentrations are determined through sample collection and immediate measurement using the Indigo blue method. The ozone feed gas is fed through a diffuser in the first (transfer) column of the two-column system and then passes through the second (contact) column where samples are collected from sample ports for both ozone and TBT analysis. Destruction of TBT with ozone or oxidation by-products is believed to occur via successive debutylation (Equation 1).



In addition to the direct oxidation of TBT by ozone, TBT can be oxidized through the formation of hydroxyl radicals during the decomposition of ozone. Reactions involving hydroxyl radical reactions are typically faster than ozone and the production of hydroxyl radicals can increase the rate at which a target compound is oxidized. The addition of hydrogen peroxide to water prior to ozonation is known to promote ozone decomposition and hydroxyl radical formation and thus has the potential to improve the process for the removal of target organic compounds.

The results of ozone and ozone + hydrogen peroxide treatment in this report (as in previous quarterly reports) are for influent waters that were created in the laboratory. These laboratory-generated waters to which TBT has been spiked to various concentrations have been utilized to date so that the reactions of ozone and ozone + peroxide are primarily with TBT and not with other non-target compounds. This approach has been favored so that the mechanisms of the treatment process can be better

¹ PCI-Wedeco, Inc.

² Model H1, INUSA, Inc.

understood after which treatment with shipyard wash waters will be conducted. All waters were prepared by passing potable (tap) water through a GAC column (>5 minutes contact time) to remove chloramines and to lower the DOC concentration in solution. These waters were collected in three 50-gallon tanks to which TBT-chloride was added to reach a desired influent level. TBT analyses of samples collected when peroxide was being added were conducted by Dr. Michael Unger of the Virginia Institute of Marine Sciences. Analysis of these samples was conducted using a solvent extraction, Grignard reagent derivitization method that was previously shown to be capable of accurate TBT analysis in the presence of an oxidant-quenching chemical added to samples immediately after collection. Quenching of oxidants that are present in a sample is imperative so that TBT does not continue to degrade in the sample bottle after collection and give inaccurate information with regard to the concentration at the time of sampling. Efforts with the TBT-hydride generation method had previously shown a consistent negative bias that was not observed in the Unger/VIMS method.

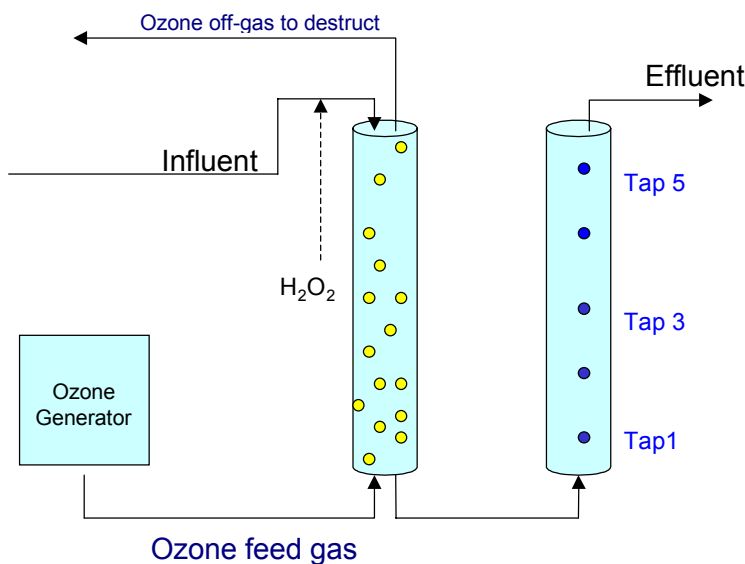


Figure 1. Schematic of laboratory ozone contacting system.

During these efforts samples were collected from taps 1 through 5 to examine the time dependent change in TBT concentrations that occur under different operating conditions and to define kinetic parameters of TBT oxidation. The relatively simple matrix simplifies the interpretation of results by isolating TBT reaction with ozone or other oxidation by-products from competing reactions. Additional information concerning the ozone system, its operation, and flow characteristics of the system are available elsewhere³.

³ Schafran, G.C., May 15, 2002 Quarterly Progress Report for USEPA Grant S-82874601-1, Evaluate Pilot and Full-Scale Treatment Processes to Remove TBT from Industrial Wastewater.

Influence of Temperature on Ozone Decomposition of TBT

Previous efforts with ozone have shown the capability of ozone to degrade TBT to very low levels particularly at warmer (~22 °C) conditions. The influence of colder temperatures was not previously determined but it was expected that a negative correlation between temperature and TBT degradation would be observed as it was for UV oxidation of TBT. Since treatment of shipyard waters will occur under cold weather conditions, it is important to understand the potential impacts that cold water conditions will have on degradation of TBT.

To conduct the cold-water TBT ozonation studies, the ozone columns and one holding tank were wrapped with insulation to minimize warming that would occur as water passed through the system. Subsequent to insulating the system influent waters were prepared by adding dechloraminated ice to dechloraminated tap water in proportions to obtain the desired influent temperature. Treatment was conducted at four different temperatures (7, 10, 15, and 25 °C), at an influent flow rate of 1.9 L/min, and at an influent TBT concentration of 3600 ng/L and pH 7.2.

Degradation of TBT by ozone was clearly impacted by temperature with effluent TBT concentrations dropping from below detection (< 15 ng/L) at 25 C to 620 ng/L at 7 C (Figure 2). At every tap it is apparent that lower temperature results in higher TBT concentrations indicating either a slowing of the direct reaction of ozone with TBT or alteration of the reaction pathway (e.g. formation of hydroxyl radicals that react with TBT) resulting in a lower rate of TBT decomposition. Ozone concentrations measured at the same locations as TBT illustrate that ozone decomposition was similarly lower at lower temperatures which by itself does not indicate which reaction mechanism might be affected (Figure 3).

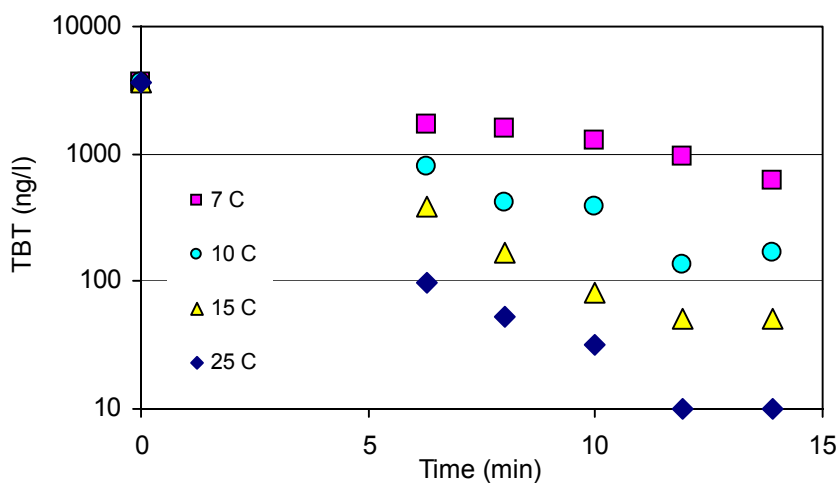


Figure 2. TBT concentrations in the contact column (taps 1-5) as a function of temperature.

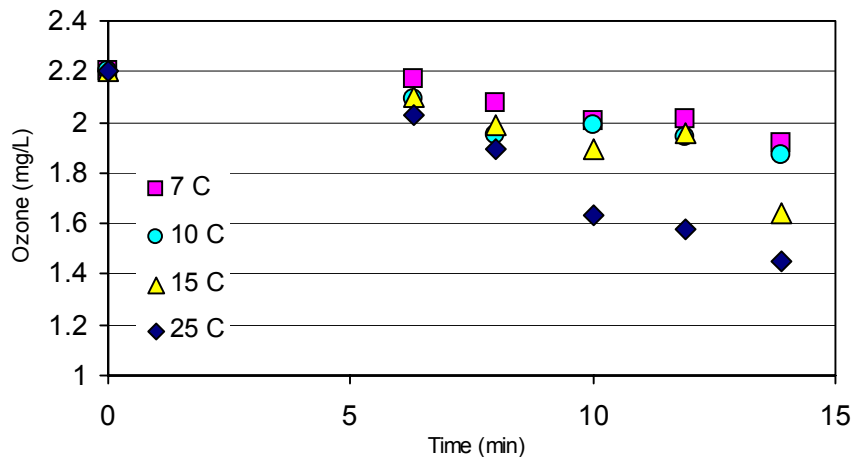


Figure 3. Ozone concentrations in the contact column (taps 1-5) as a function of temperature. Time zero ozone concentration is calculated based on assumption of 100% ozone transfer.

Influence of Hydrogen Peroxide on Decomposition of TBT by Ozone

Use of hydrogen peroxide has been reported in many studies to assist in the formation of hydroxyl radicals and increase the rate at which target organic compounds are decomposed. Since it appears that cold water conditions might substantially impact the ability to reach the desired TBT concentrations (i.e. < regulatory limit), the ability of peroxide to improve TBT removal under “cold” water conditions was particularly of interest. Two sets of experiments were run under warm and cold water conditions to evaluate how TBT decomposition would be influenced by peroxide addition. The results of these experiments are included below.

Evaluation of the influence of hydrogen peroxide on TBT removal under cold water conditions was conducted at 6-7 °C at an ozone dose of 2.2 mg/L and an influent TBT concentration of 3,600 ng/L. Two runs were conducted with one at 0 mg/L and the second with a dose of 1.74 mg/L peroxide. The dose of 1.74 mg/L of hydrogen peroxide was selected based on a number of previous studies indicating an optimum molar ratio of 0.5 H₂O₂/O₃. Under cold water conditions concentrations of TBT were substantially lower with the addition of peroxide compared to similar treatment conditions without peroxide (Figure 4). The peroxide-dosed water exhibited greater removal of TBT but with the reaction essentially going to completion by the first sampling tap with no additional removal occurring with additional contact time in the second column. This trend suggests that ozone and any radicals that were formed were consumed before the first tap and were the limiting factor in no additional TBT removal. Evaluation of ozone concentrations for the two peroxide conditions illustrates that ozone was indeed completely consumed by the first tap in the peroxide-dosed water but that ozone was plentiful in the water without peroxide and continued to oxidize TBT (Figure 5).

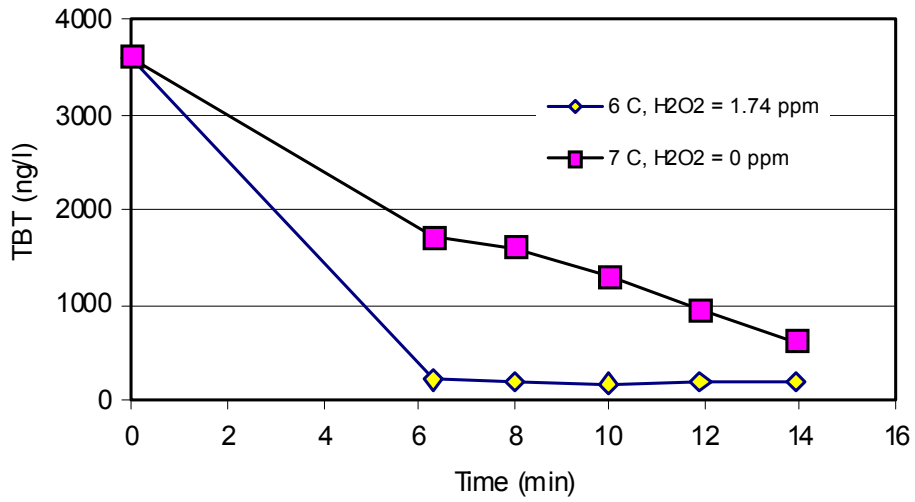


Figure 4. TBT concentrations in the contact column (taps 1-5) as a function of hydrogen peroxide dose under cold water conditions.

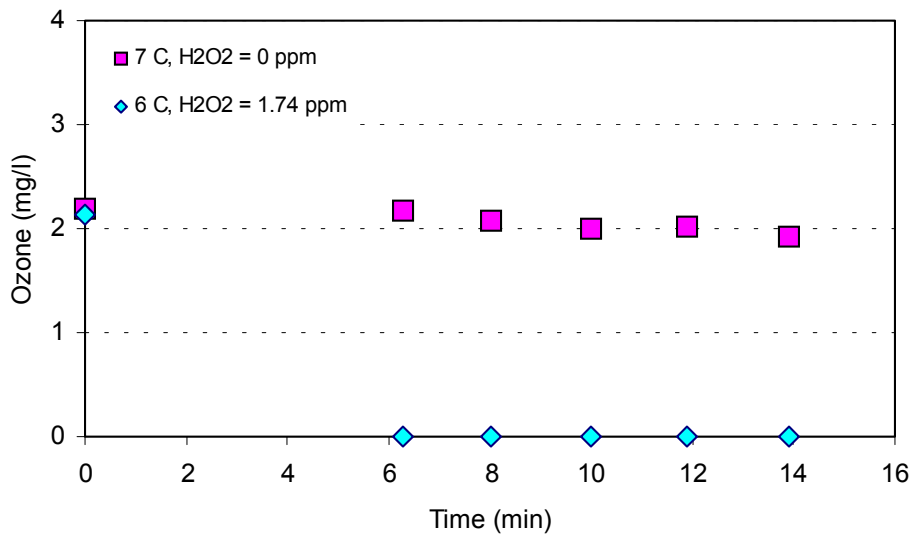


Figure 5. Ozone concentrations in the contact column (taps 1-5) as a function of hydrogen peroxide concentration.

The cold water peroxide-dosed treatment results clearly illustrate an improvement in treatment due to hydrogen peroxide addition and suggest that additional ozone or possibly less peroxide would have provided even greater removal of TBT. Hydrogen peroxide concentrations at four of the five sample taps was observed to be 1.3 mg/L indicating that only 0.4 mg/L of peroxide was consumed during treatment and that additional peroxide was available to react with ozone. As TBT concentrations did not exhibit a reduction at taps 2 through 5 (and only a minor decrease between taps 1 and 2) it can be surmised that under the conditions of the experiment that peroxide is not a strong, direct oxidizer of TBT. However, it does react rapidly with ozone even under cold water conditions.

Ozone + peroxide study efforts were also conducted at warm water conditions (laboratory ambient temperature of approximately 25 C) under three hydrogen peroxide concentrations; 0, 0.5, and 1.0 mg/L H₂O₂. Operational conditions (O₃ dose = 2.2 mg/L, water flow = 1.9 L/min, pH = 6.8) were consistent with previous efforts but influent concentrations of TBT were increased to 9,200 ng/L in anticipation of rapid TBT decomposition with the addition of peroxide under warm water conditions. Peroxide concentrations were also lowered in these efforts since it was suspected that excess peroxide may have inhibited additional TBT decomposition in the cold water studies.

The warm water treatment efforts provided results that were the converse of the cold water conditions in terms of the benefit of peroxide addition to TBT removal (Figure 6). The addition of peroxide resulted in higher TBT concentrations at all sample taps compared to no peroxide addition and the higher peroxide dose provided the poorest level of treatment. These results are divergent from the observations at 6-7 °C where it was observed that TBT concentrations were lowered and treatment was improved with the addition of peroxide. Similar to the observations under cold water conditions, ozone was completely decomposed in the first reactor with no measurable ozone concentration occurring at any of the sample taps in the second column (Figure 7). Hydrogen peroxide was also present (0.9 mg/L in the 1.0 mg/L dosed water and 0.34 mg/L in the 0.5 mg/L dosed water) through the second contactor column indicating it was present in excess of the amount needed to promote hydroxyl radical formation.

The warm water findings with hydrogen peroxide are particularly important in the research efforts evaluating ozone + peroxide as a treatment option for the removal of TBT from shipyard washwaters since it indicates that excess hydrogen peroxide even at low concentrations can negatively impact TBT removal instead of improving it. This effect is likely due to peroxide reaction with hydroxyl radicals formed during reaction with ozone as illustrated in Figure 8. In this figure, there are three concurrent “terminal” reactions for ozone. All three reactions proceed concurrently but at different rates depending on solution conditions. With increasing concentrations of peroxide added to a solution the reactions will shift from a dominance of reaction 1 (direct oxidation of TBT by ozone), to reaction 2 (hydroxyl radical oxidation of TBT), to reaction 3 (hydroxyl radical consumed by peroxide without reaction with TBT). Reaction 3 appears to have dominated during the warm water studies and the effect of increasing the peroxide concentration from 0.5 to 1.0 mg/L resulted in even less TBT oxidation.

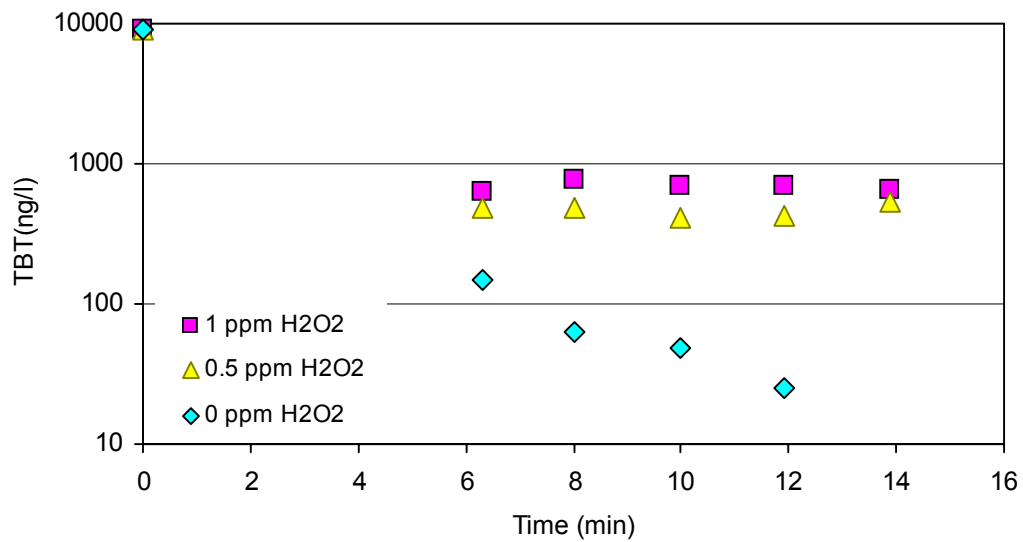


Figure 6. TBT concentrations in the contact column (taps 1-5) as a function of hydrogen peroxide dose under warm water conditions.

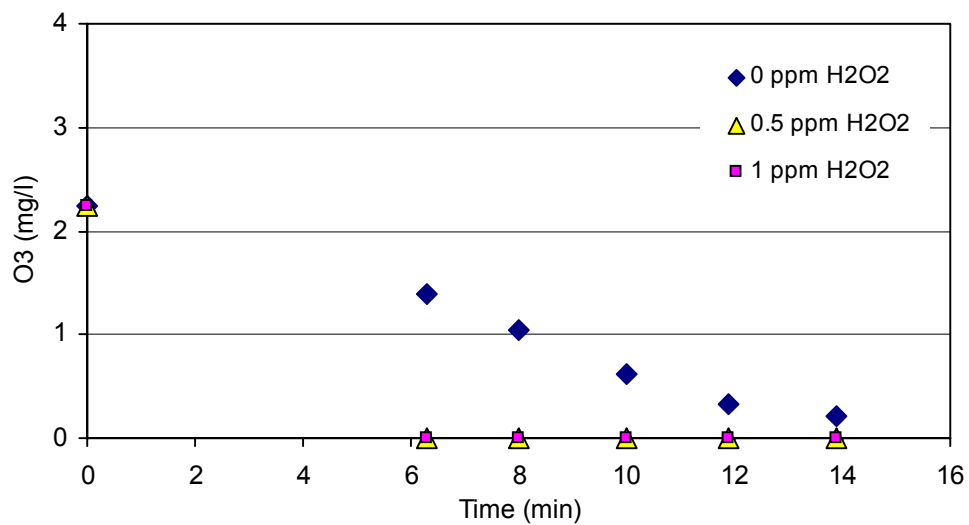


Figure 7. Ozone concentrations in the contact column (taps 1-5) as a function of hydrogen peroxide concentration under warm water conditions.

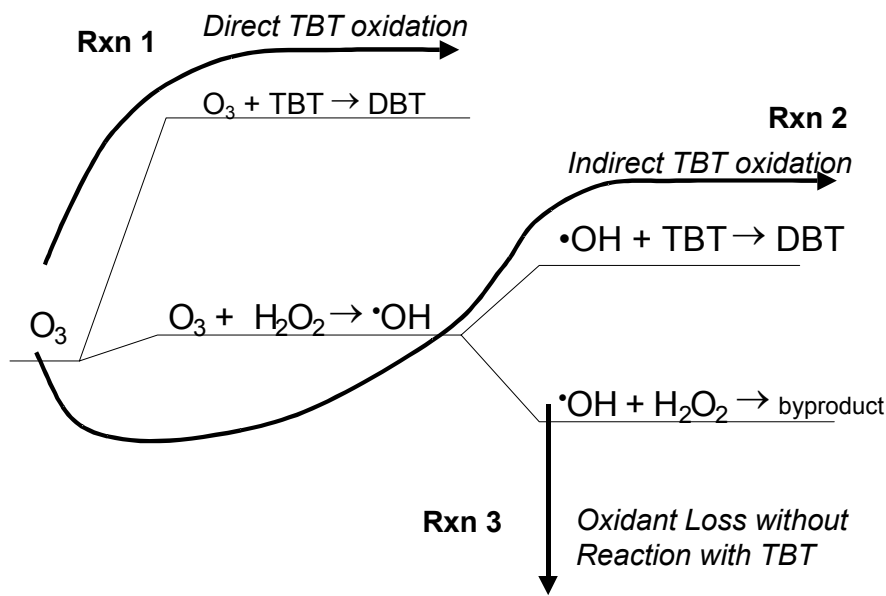


Figure 8. Simplified representation of reaction pathways involving TBT oxidation during treatment with ozone and peroxide.

Ozone Consuming Reactions

Oxidation reactions in water can be complex and involve many potential interactions that do not involve the compound(s) targeted for removal. In the case of ozone oxidation of TBT, any other substance present in water that reacts with ozone in the time frame (<15 minutes) in which TBT is oxidized can interfere with the removal of TBT. If ozone is to be incorporated as a unit process in the treatment of shipyard wash waters then it will be integrated as the final unit process of the current process configuration. This effort would place the ozone system following the activated carbon units resulting in the lowest particle and organic matter concentrations entering the ozone system. While particle and organic matter concentrations will be much reduced, they will still be present and exert some oxidant demand. Efforts to examine the oxidant demand associated with particles and organic matter were conducted to better understand the contributions of each to ozone demand.

In previous study efforts the presence of particulate material in the final effluent of the full-scale treatment plant has been identified as a contributing factor to TBT concentrations present in the effluent above the Virginia discharge limit. Sources of this particulate material have been examined and it appears that the GAC fines may mobilize from contactor columns and contribute to the particulate concentration of the final effluent. Since GAC fines are a potential source of particulate material to the final effluent, a set of experiments was conducted to examine the contribution of GAC fines to ozone demand and its impact on TBT removal. This effort was conducted using tap

water that was passed through a GAC column (to dechloraminate the water) and then filtered to various degrees (no filtration, 1 μm and 0.45 μm filtration) to examine the potential contribution of GAC fines to ozone demand. The ozone system was run under typical conditions (described previously) and ozone concentrations were measured at the five sample taps on the second column. Ozone concentrations for the unfiltered and 1 μm -filtered waters was 2.2 mg/L while the 0.45 μm -filtered water was slightly lower at 1.9 mg/L. TBT spiked in the makeup water (influent) was 3100 ng/L for the unfiltered and 1 μm -filtered water while no TBT was added to the 0.45 μm -filtered water.

Ozone concentrations were affected by the water that was used and clearly illustrate a difference in ozone demand (i.e. consumed) with degree of filtration. The unfiltered water that was collected directly from the effluent of the laboratory GAC column exhibited the highest ozone demand of the three waters examined with the ozone demand decreasing with the pore size of the filter used for filtration (Figure 9). It should be noted that while no TBT was added to the 0.45 μm -filtered water and this could have contributed to the lower ozone demand observed, the decreased demand is consistent and believed to be the result of lower particle concentrations in the influent water. It is also important to note that all waters had what would be considered low suspended solids concentrations measuring below detection (~ 1 mg/L) in the effluent of the unfiltered water.

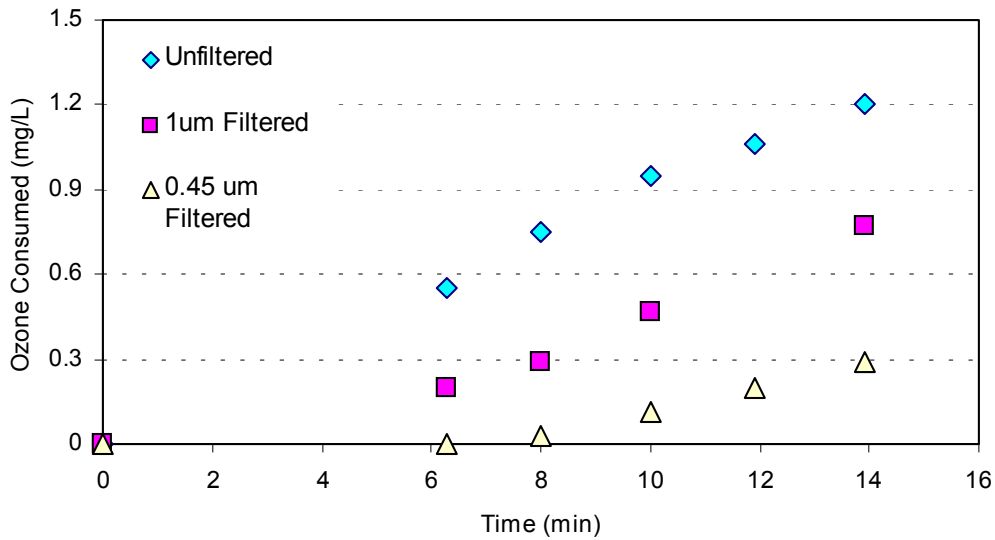


Figure 9. Variation in ozone demand as a function of source water. Ozone demand = ozone dose – ozone residual.

Concentrations of TBT were also measured in samples collected during treatment of the unfiltered and 1 μm -filtered waters. In both of the se waters the tap 5 samples were below detection and the tap 1 samples were below 60 ng/L. The results indicate that TBT removal was not adversely affected by the presence of particulate material even though ozone demand was increased by its presence. That TBT removal was not adversely affected may be the result of adequate ozone concentrations present throughout the ozone system ($\geq 1 \text{ mg O}_3/\text{L}$) providing enough oxidation to overcome the additional demand exerted by the particulate material. This observation is important since it suggests that if the ozone demand not associated with TBT is overcome, then adequate oxidation of TBT may be achieved.

Organic matter is another source of ozone demand and efforts to evaluate its impact on TBT treatment were studied under carefully controlled conditions. Organic matter varies widely in composition and reactivity in natural and industrial waters with aromatic organic compounds being particularly reactive with ozone. In the studies conducted here Aldrich humic acid, an organic matter source rich in aromatic material, was used as the source material. Dechloraminated tap water (0.6 mg/L dissolved organic carbon, DOC) was run directly and spiked to two different concentrations (measured concentrations of 2.6 and 6.8 mg/L DOC) in these efforts. Influent TBT concentration for all three runs was 3600 ng/L.

The influence of the organic matter concentrations on ozone reaction with TBT was striking and clearly increasing concentrations of DOC were detrimental to the removal of TBT (Figure 10). TBT concentrations in the water that was not spiked with DOC (background DOC = 0.6 mg/L) were similar to the previous efforts when water of this type. These data suggest that most of the available ozone was consumed in reactions with the humic acid that was added and thus not available to react with the TBT in solution. Ozone measurements in the second column were all below detection (Figure 11) for the organic matter-spiked waters indicating that the ozone that was transferred into solution in the first column was consumed quickly and provided no additional TBT removal in the second column.

While treatment performance was poor for the organic matter-spiked waters, it should be recognized that the ozone dose was relatively low (2.2 mg/L) and the organic matter reactivity likely exceeded that of the organic matter normally present in the TBT-containing wash waters. It would be expected that with an increase in ozone dose measurable ozone residual in the second column would occur and removal of TBT to levels similar to that of the unspiked (0.6 mg/L DOC) TBT water would occur.

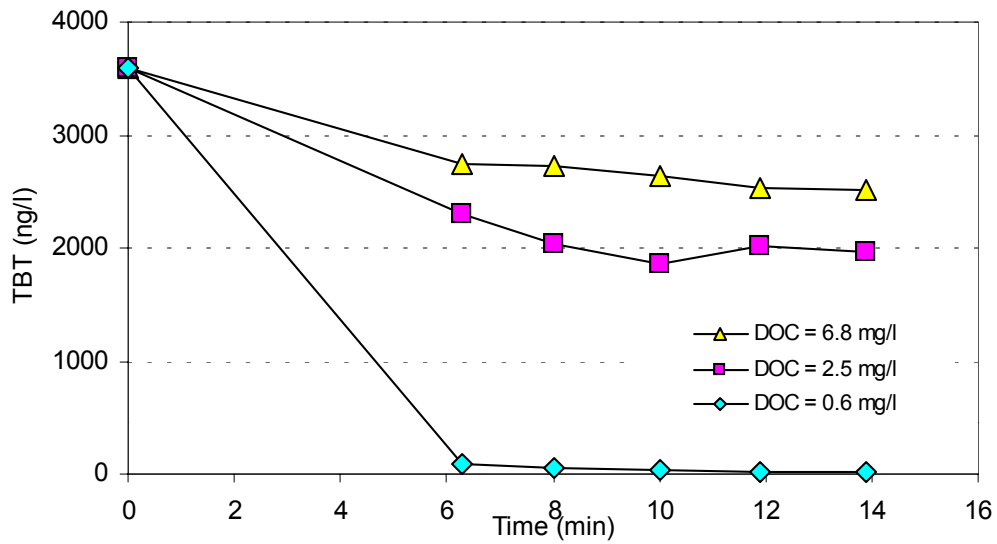


Figure 10. TBT concentrations in the influent and contact column of the laboratory ozone system as a function of organic matter (DOC) concentration.

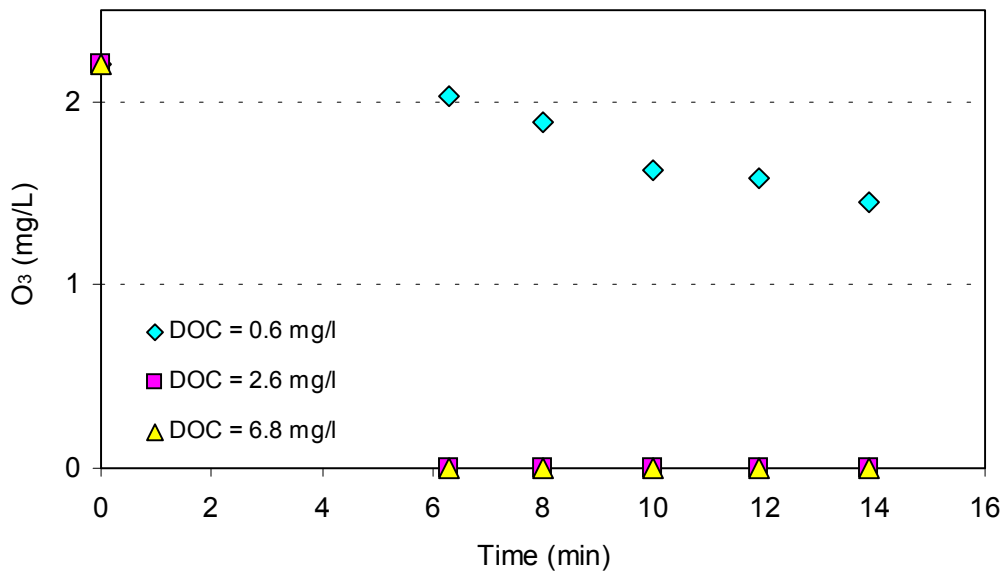


Figure 11. Ozone concentrations across the pilot ozone system as a function of organic matter (DOC) concentration.

Removal of Copper and Zinc Utilizing Activated Carbon

In evaluating the removal of copper and zinc during operation of the full-scale TBT treatment plant, evidence of copper and zinc removal in the activated carbon columns was observed. Copper removal was observed to occur across a wide range of influent concentrations and appeared to be irreversible under the operating conditions of the treatment plant. Activated carbon removal of zinc was less effective and evidence indicated that frequently zinc was conservatively transported through the activated carbon columns (i.e. no removal) or was mobilized and was present in the effluent at higher concentrations than the influent. Understanding the mechanisms of adsorption for copper and zinc on activated carbon are particularly important since it appears that changes to the waste stream composition could potentially cause sudden releases of these metals. If numeric limits for either of these metals is established in a discharge permit, periodic release from the adsorbent could lead to conditions where permit limits could be exceeded.

To better understand the removal of copper and zinc onto activated carbon, a laboratory-based continuous-flow adsorption study was conducted with goals to identify the adsorption capacity for copper and zinc and to examine the fate of adsorbed metal under different treatment conditions. The study was conducted using a 2.5 cm diameter column containing 19.3 grams of Calgon F-400 carbon (Figure 12). Water adjusted to pH 6.5 to 6.7 and containing 1,000 ppb copper and zinc from AAS stock solutions and 0.05 M NaNO₃ was pumped through the system at a rate of 5.5 mL/min providing an empty bed contact time of 6.4 minutes. The column was typically operated on a daily basis and shut down during the evenings. Effluent samples were collected every 0.55 liters and analyzed for copper, zinc, and effluent pH.



Figure 12. Laboratory GAC contact column system for Cu and Zn removal studies.

Copper and zinc removal by activated carbon exhibited distinctly different trends during the column adsorption study (Figure 13). Removal of both metals was nearly 100% initially but declined rapidly for zinc. At the 257-bed volume mark, the influent solution was mistakenly made up without the background electrolyte (0.05 M NaNO₃) which caused a dramatic change in the performance of the column. Zinc concentrations increased from 80% of the influent concentration to over 140% indicating that previously removed zinc was remobilized to the column effluent. Copper concentrations were also significantly affected as evidenced by removal reduced from nearly 100% to less than 50%. Upon use of a new batch of influent with the 0.05 M NaNO₃ added, copper removal returned to near 100% removal while zinc removal returned to a point of higher removal (lower effluent concentration) than observed at the time of the errant influent solution. This level of removal quickly diminished and removal was no longer observed after 650 bed volumes of water had been treated.

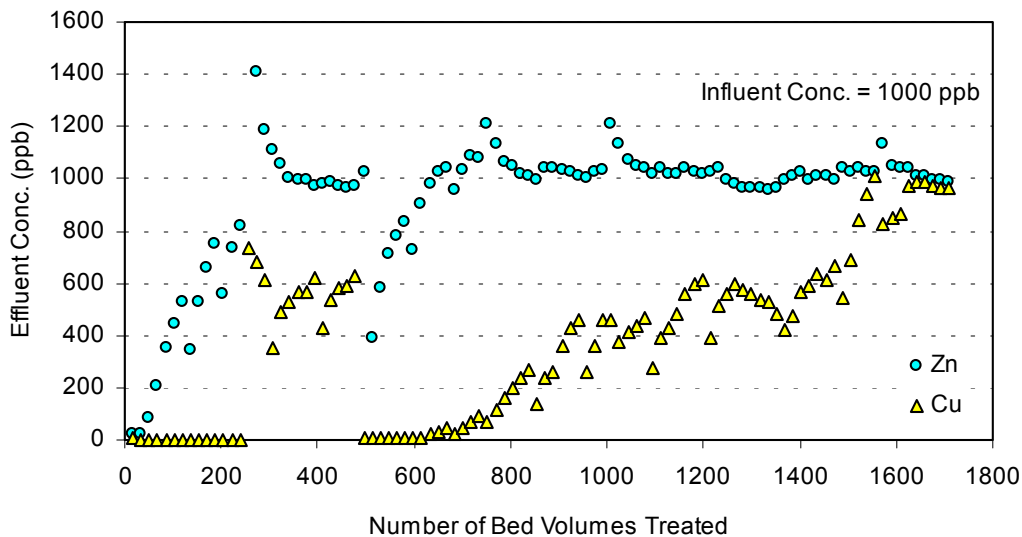


Figure 13. Laboratory GAC contact column system for copper and zinc removal studies.

At approximately the time that zinc removal was no longer observed, copper removal began to decrease in the manner typically observed for a breakthrough curve. The increasing effluent concentration however was dampened and removal was extended possibly due to the intermittent (similar to full-scale treatment) operation of the column. Exhaustion of the column for copper removal was reached at approximately 1600 bed volumes. At this point, zinc saturation was determined to be 0.17 mg Zn/g GAC while saturation of copper was calculated as 1.72 mg Cu/g GAC. Consequently, it can be seen

that for the GAC tested and under the conditions of this effort, the carbon had a 10-fold higher capacity for copper than zinc.

Both copper and zinc exhibited trends that indicate that periodic operation of the column might affect metal removal and a change in influent composition could cause a dramatic increase in effluent metal concentration in response. It was observed that at the beginning of a day's run, zinc concentrations were often at the highest level and decreased with treatment duration. Conversely, copper concentrations were typically lowest at the beginning of a day and increased with run time. This trend signifies the variation in concentration that can occur over a relatively short time interval (hours) and how there might be little advance warning before concentrations would go from within compliance (assuming numeric limits in the low ppb) to out of compliance concentrations. One caveat in interpreting the trends and observations of this study is that the influent solution was free of organic matter to evaluate the difference in metal adsorption due to adsorption of each metal in an inorganic form. The presence of organic matter in solution is expected to increase the adsorption of these metals and is being studied in a follow on experiment.

TBT Removal by Ultraviolet Irradiation

Previous study efforts with a laboratory-scale reactor and in a single field study at Norfolk Naval Shipyard (NNSY) have illustrated the capability of ultraviolet (UV) irradiation to decompose TBT to low pptr levels. In the field study at NNSY concentrations were lowered from 14,000 pptr, a concentration more than triple the highest concentration ever observed in the effluent of the full-scale treatment plant, to less than 100 pptr. A substantial benefit to the removal of TBT that was proven at NNSY was the addition of hydrogen peroxide. Laboratory study efforts presented here concentrated on confirming the benefit of peroxide addition and the effect of dosage of peroxide. All samples for this study effort were analyzed in the laboratory of Dr. Michael Unger, Virginia Institute of Marine Sciences.

TBT removal was examined using the laboratory batch reactor evaluating various peroxide concentrations (0, 75, and 145 mg/L) and lamp intensity (10 lamps and 16 lamps). An influent water was made up to 2600 ng/L in deionized, distilled water and then used for all experiments reported here. Each data point was generated using a separate aliquot of water that was irradiated for the indicated period of time and with the indicated dose of hydrogen peroxide. At the end of the irradiation period the lamps were shut off, a small aliquot of sample was removed for measurement of peroxide and then the peroxide-dosed samples were quenched with a reducing agent (catalyse).

Results of this effort confirm that peroxide provides a considerable increase to the rate of TBT oxidation and that improved TBT removal was not sensitive to peroxide concentration at the levels tested (Figures 14 and 15). As observed previously, the first-

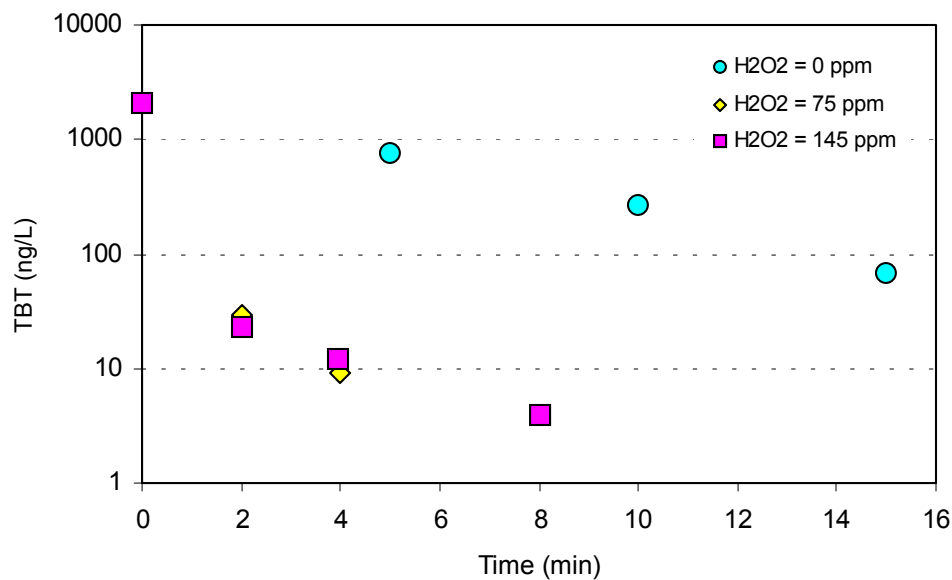


Figure 14. TBT concentrations as a function of UV irradiation time and hydrogen peroxide concentration. UV irradiation conducted with 16 lamps.

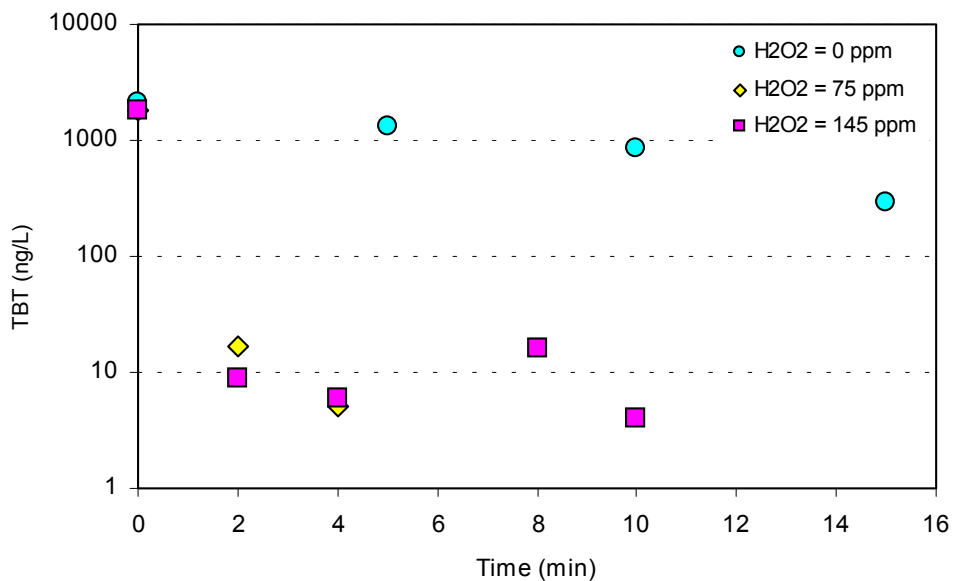


Figure 15. TBT concentrations as a function of UV irradiation time and hydrogen peroxide concentration. UV irradiation conducted with 10 lamps.

order rate constant for TBT removal in the absence of peroxide was proportional to the number of lamps used ($k_{16} = 0.23 \text{ min}^{-1}$; $k_{10} = 0.13 \text{ min}^{-1}$). However, little difference was observed in the degradation of TBT between the 16-lamp and 10-lamp effort when using peroxide. This apparent effect may be directly attributable to the very high rate of decomposition that lowered the TBT concentrations to less than 30 ng/L in two minutes of irradiation time for both the 10-lamp and 16-lamp efforts. Note, “missing” TBT values at greater than four minutes reaction time are actually concentrations that were analytically determined to be below detection and not plotted. Hydrogen peroxide concentrations measured during these efforts illustrate that peroxide was available throughout the period of irradiation (Figure 16).

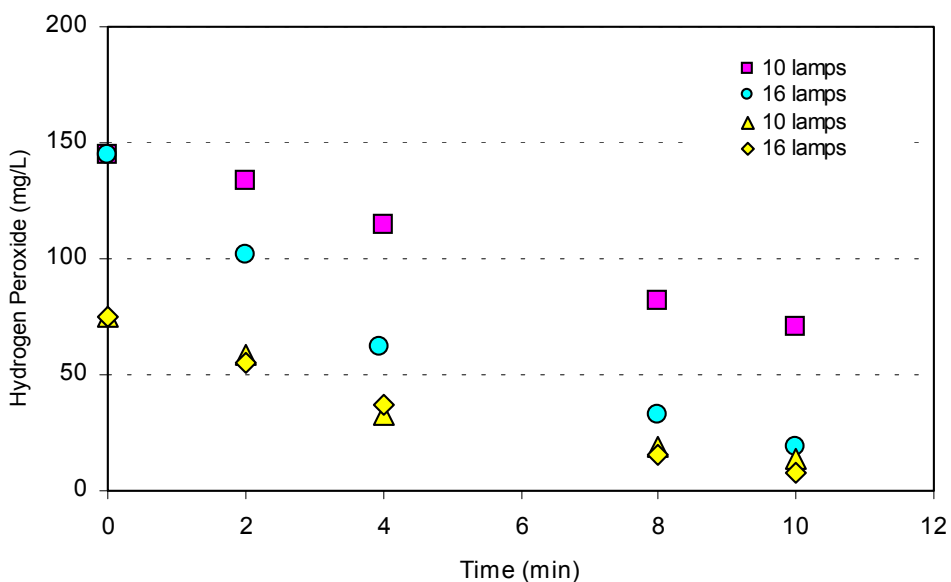


Figure 16. Hydrogen peroxide concentrations as a function of UV irradiation time and number of lamps. Initial concentrations are observable at time = 0 minutes.

Summary and Future Efforts

Oxidation of TBT using ozone has been shown capable of removing TBT to concentrations below detection under warm water conditions but under cold weather conditions the rate of decomposition is severely reduced and in efforts reported here exceeded the 50 ng/L discharge limit. Efforts to increase the rate of TBT oxidation with ozone by adding peroxide to stimulate hydroxyl radical formation were observed under cold water conditions but under warm water conditions, peroxide addition worsened TBT removal compared to no peroxide addition. These results indicate that hydroxyl radical

formation and oxidation of TBT is very sensitive to peroxide concentration and would likely be difficult without sophisticated and continuous (real time) monitoring of ozone concentrations to maximize TBT removal.

Copper and zinc removal by activated carbon were observed to occur when they are primarily in inorganic forms with the capacity of activated carbon being 10-fold higher for copper than zinc under neutral pH conditions. The fate of adsorbed metals may not be permanent immobilization as an inadvertent change in the background electrolyte was shown to cause previously removed zinc to desorb causing effluent concentrations to exceed the influent concentrations. Work is continuing to examine the sensitivity of the adsorbed metals to remobilization with changes in influent composition. In addition, laboratory experimental efforts with waters containing organic matter (both synthetic and actual shipyard wash waters) will be studied to determine the fate of the metals and the ability of activated carbon for potential control of metal discharges to the environment.

UV irradiation studies with peroxide confirm the capability of peroxide-enhanced oxidation of TBT and the relative insensitivity to peroxide concentration. Efforts are continuing using real wash water samples in laboratory studies and preparations are underway for another full-scale evaluation at Norfolk Naval Shipyard using their full-scale UV system.

Findings of research supported under this grant and previous grants will be presented at the Society of Naval Architects and Marine Engineers (SNAME) Ship Production Symposium on September 25, 2002 in Boston, MA and published in the proceedings. A copy of this paper is included as an attachment to this report.