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D. E. Maier, *Kansas State University*

C. A. Campabadal, *Purdue University*

C. P. Woloshuk, *Purdue University*

L. Mason, *Purdue University*



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## Continuous ozonation treatment systems as other alternative more efficient grain protection technologies

Maier, D.E.\*<sup>1</sup>, Campabadal, C.A.#<sup>2</sup>, Woloshuk, C.P.<sup>3</sup>, Mason, L.<sup>4</sup>

<sup>1</sup> Department of Grain Science and Industry, Kansas State University, 201 Shellenberger Hall, Manhattan, KS 66506, Email: dmaier@k-state.edu

<sup>2</sup> Agricultural and Biological Engineering, Purdue University, 225 South University St. West Lafayette, Indiana, USA 47906-2093

<sup>3</sup> Botany & Plant Pathology Department, Purdue University, 1155 LILY, West Lafayette, IN 47907-1155

<sup>4</sup> Department of Entomology, Purdue University, 1158 SMTH, West Lafayette, IN 47907-1158

\* Corresponding Author

# Presenting Author

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### Abstract

Previous static bed ozonation scale-up and demonstration trials have proven the use of ozone as an effective technology for grain protection without affecting its end-use quality. Due to the lack of current availability of high capacity ozone producing generators, grain treatment through static bed ozonation systems are limited to be used in metal silos of capacities smaller than 644-t. Also, the trials have shown that treatment time has to be of no less than 4 d during application in order to be effective for pest control. Therefore, more efficient ozonation treatment systems are needed for proper ozone usage for stored product protection. The primary objective of these research studies was to design and test a semi-continuous counter-flow ozonation and a continuous ozonation flow treatment systems in order to ozonate grain at faster rates based on the concentration-time product (CTP) of ozone required to achieve 100% insect mortality and effective mold reduction in grain. The procedure of the counter-flow semi-continuous ozonation system consisted of removing each grain layer inside a metal silo with a tapered unloading auger after each layer reached the desired ozone CTP. The treated grain is subsequently transported to a storage or shipping silo. The continuous ozonation flow system involves applying high ozone concentrations through a modified grain loading screw conveyor where ozone and grain are moving continuously in the same flow direction. The counter-flow semi-continuous ozonation system was successfully tested and proved to be a technically feasible tool for pest control and mold reduction. The continuous ozonation flow system was proved as an effective tool for treating grain during handling while achieving 100% insect mortality, and effective mold reduction.

Keywords: Ozonation, Continuous treatment, Grain, Pest control, Molds.

### 1. Introduction

Previous fixed bed ozonation scale-up and demonstration trials have been conducted in grain storage structures and have proven the use of ozone as an effective technology for grain protection without affecting its end-use quality. This fixed bed ozonation system aimed at achieving an initial sterilization phase by reaching a target concentration of 50 ppm at the opposite side of the ozone inlet and maintaining it for 72 h. Ozone is artificially produced by a generator and introduced to the grain storage structure at a constant rate. For this treatment setup, usually a recirculation or exhausting system is needed so ozone is not wasted once the leading front exits the grain mass and is not just exhausted into the environment. Among the advantages of ozonation compared to traditional phosphine fumigation for pest control are that it can be generated at the treatment site and no residue is left on the treated product (Strait, 1998). Due to the lack of current commercial availability of high capacity ozone producing generators, grain treatment with ozone through fixed bed systems are limited to be used in metal grain silos of capacities smaller than 644-t. Also, the trials have shown that treatment time has to be of no less than 4 d during application in order to be effective for pest control. Therefore, more efficient ozonation treatment systems are needed for proper ozone usage for stored product protection. The ozonation treatment alternatives were built on knowledge gained from previous fixed bed ozonation trials and focused on the development of a semi-continuous counterflow ozonation system and a continuous ozonation flow system in order to treat grain for fungal spores sterilization and insect eradication at a

continuous rate that will result in a faster treatment time compared to the current fixed bed treatment method.

The semi-continuous counterflow ozonation system was designed based on an in-bin continuous flow dryer system used for on-farm grain drying in metal round silos (Marks et al., 1993). These drying systems have tapered unloading augers in the grain silo floor that are capable of unloading the silo's bottom grain layer which can result in a removal of a depth of 15.2 to 30.4 cm of grain in the silo after completion of one full revolution. The procedure for the semi-continuous counterflow ozonation treatment involves removing the bottom grain layer after it reached the desired ozone concentration to achieve insect mortality, mold reduction, and/or off-odor removal. The treated grain is subsequently transported to a semi-truck trailer, grain wagon or second storage silo. At the same time, untreated grain from another storage silo is added on top of the grain surface in the same amount as the treated grain that was removed by the tapered unloading auger in order to maintain a constant grain depth during the treatment process. The system control variables are; airflow, ozone mass flow and exposure time. Airflow is controlled by a variable speed fan that is sized up to deliver at least the minimum air velocity of 0.03 m/s to move ozone through a grain mass (Mendez et al., 2002) and a maximum that will not cause too great of an ozone dilution effect in the bottom grain layers. The ozone mass flow is controlled by the ozone producing generator. The only safe current method to control this variable is by shutting down one or more of the four chambers of the ozone generator. The exposure time of the grain to ozone is controlled by determining the CT product as a function of the ozone concentration at each grain depth needed for the desired treatment effect (odor removal, fungi sterilization and insect mortality). Once the desired CT product is achieved and maintained, the unloading tapered auger is programmed to remove the bottom treated grain layer.

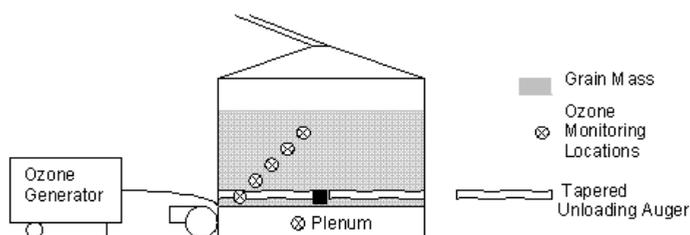
The continuous ozonation flow system consists in applying high ozone concentrations through a modified screw conveyor in the same flow direction that the grain is moving continuously during a handling operation in order to achieve the desired CT product for insect eradication. The treated grain at the end of the modified grain loading screw conveyor falls through a flow directed pipe into a receiving chamber. At the same time, untreated grain is continuously moved into the modified screw conveyor at a constant rate. The control variables of the system are: grain residence time and ozone mass flow. The grain residence time is controlled by the rotational speed of the modified screw conveyor auger which is defined as the time it takes the first kernel introduced into the modified screw conveyor to come out at the other end after it has been treated with ozone. The ozone mass flow is controlled in the same way as in the semi-continuous counterflow ozonation system. The primary objective of these research studies was to design and test a semi-continuous counter-flow ozonation based on achieving a constant concentration-time product CTP throughout the treatment in the bottom layer of the grain mass inside the silo and to quantify any possible mold reduction and to design and test a continuous ozonation flow treatment system in order to ozonate grain at faster rates based on the CTP of ozone required to achieve 100% insect mortality and effective mold reduction in grain.

## *2. Materials and methods*

In both semi-continuous counterflow and continuous ozonation flow treatment trials, ozone was produced using the corona discharge technology by a four chamber generator (OzoBlast) manufactured by O3Co. (Aberdeen, ID) that has a rated capacity of producing 1,000 g/h.

### *2.1. Semi-continuous counterflow ozonation treatment trial*

The semi-continuous counterflow ozonation treatment was conducted in a corrugated metal grain silo (Fig. 1) of a capacity of 250 t with the following characteristics of 9.1 m diameter with a sidewall height of 6.7 m and a centrifugal fan powered by a 2.2 kW motor, a propane burner, a grain distributor, and a tapered sweep unloading auger system of a diameter of 9 m (Shivvers, Corydon, IA) which rotates around the silo diameter removing 12 t of grain for each full rotation. During the treatment trial, the grain drying system fan was replaced by a 0.35 m diameter and 3,500 rpm axial fan manufactured by Sukup Manufacturing Company (Sheffield, IA) powered by a 0.75 kW motor that was controlled by a variable frequency drive (VFD) in order to control the airflow that pushed the ozone into the grain mass. The airflow values produced by the axial fan were quantified by previously measuring them using a fan performance test (ANSI/AMCA 210, 1999) for different fan rotational velocities (rpm) and at different fixed pressures.



**Figure 1** Semi-continuous counterflow ozonation treatment system in dryeration grain silo with ozone monitoring setup.

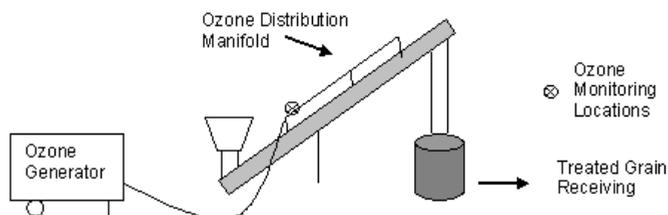
Ozone concentration was quantified with a monitoring system that measured the concentration at different depths of the grain mass (Fig. 1) using multiple monitoring lines connected to an ozone analyzer model IN-2000 made by IN USA Inc. (Boston, MA). Ozone produced by the generator was discharged from each of its four chambers out through 2.54 cm Teflon supply lines into the grain silo. The four ozone supply lines were positioned to empty into the plenum of the grain silo through the fan transition in order to achieve uniform distribution of ozone below the silo's perforated drying floor.

The trial was conducted by treating 190 t of maize in cycles of a constant grain mass of 63 t. In each treatment run the bottom grain layer of 12 t was exposed to the desired CTP. After treatment, each bottom grain layer was removed and an untreated grain layer of 12 t was added to the grain mass.

Mold colonies from each bottom grain layer inside the grain silo before and after treatment were quantified by determining the number of colony forming units (CFU) on the grain surface. Samples of 25 g of corn was added to 50 mL of 0.05% Triton X-100 solution and mixed for 2 min. The wash was serially diluted and plated onto malt salt medium. Plates were incubated at 28°C for 3 to 5 d.

## 2.2. Continuous ozonation flow treatment trial

The continuous ozonation flow system was designed as a modified grain loading screw conveyor (Fig. 2) by LynnTech (College Station, TX). It was made from stainless steel (SAE grade 316) with a length of 6.3 m and a diameter of 0.102 m. The internal shaft had a length of 6.3 m, a diameter of 0.038 m and a pitch of 0.102 m. Attached to the base of the conveyor was a hopper of a 22.7 kg capacity used to feed grain into the system. At the exit of the conveyor was a drop shoot made of 0.102 m PVC pipe ending in a collection bucket covered with a carbon fiber filter to destroy any ozone exiting the system. Along the length of the screw conveyor was a manifold that distributed the ozone through six injection points separated every 1.5 m. A 0.75 kW motor with an inverter connected through a variable frequency drive manufactured by Maraton Electric (Wausau, WI) was used to control the rotational speed of the screw conveyor. Throughout the trials, the screw conveyor was set at a fixed inclined angle of 35 degrees.



**Figure 2** Continuous ozonation flow treatment system with ozone distribution manifold and monitoring setup.

Ozone concentration in the distribution manifold was determined with an ozone analyzer model H1 manufactured by IN USA Inc. (Boston, MA). Ozone produced by the generator was discharged from each of its four chambers through 2.54 cm Teflon supply lines into a 5.08 cm Teflon main supply line that was connected to the ozone distribution manifold along the modified screw conveyor.

The trial was performed by treating 15.9 kg samples of maize at different CTP in the modified screw conveyor with dry ozone produced directly from the ozone generator and humidified ozone. The

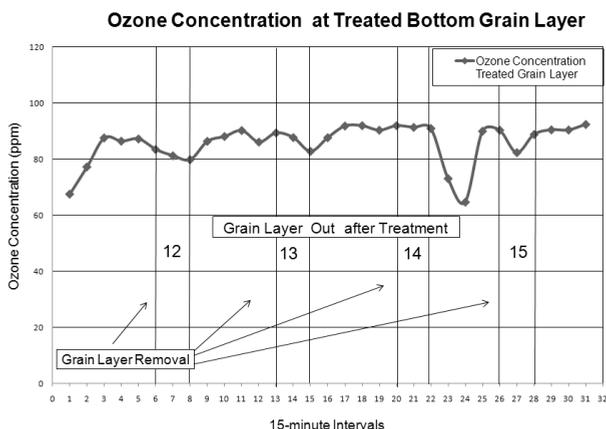
humidified ozone was obtained by producing dry ozone from the ozone generator and introducing it into a water tank to absorb moisture after it is passed through 90 L of water. The humidity level of this mixture was not determined.

Insect mortality was used as the main parameter and it was determined using five laboratory-made insect bioassays for each of the two insect species. These insects were; adults of the red flour beetle [not used at a later stage] *Tribolium castaneum* (Herbst) and the maize weevil [not used at a later stage] *Sitophilus zeamais* Motschulsky. Each insect bioassay contained 10 insects and were placed and mixed on each 15.9 kg grain samples before each treatment and control trial runs. Mold reduction was measured using the same procedure mentioned for the semi-continuous counterflow ozonation trial.

### 3. Results and discussion

#### 3.1. Semi-continuous counterflow ozonation treatment trial

During the semi-continuous counterflow ozonation trials in the bottom treated grain layer, an approximate constant ozone concentration value of 90 ppm was maintained throughout the treatment process (Fig. 3) and the treated grain layers during the full cycle had an average CTP of 425 ppm.h. Three different phases can be identified during the treatment process. The first phase involves buildup of ozone concentration during which ozone reacts with the available reactive sites in the bottom grain layer. The second phase involves maintaining a steady state of one hour between the grain and ozone at its maximum concentration. The third phase ozone concentration decreases due to unloading of the bottom grain layer. The airflow rate affected the ozone concentration during the three phases. During grain layer removal, airflow was increased to avoid ozone leakage through the unloading system. Airflow remained mainly constant through the steady state of the treatment. Ozone after it reached its maximum concentration produced a sterilizing effect in the bottom grain layer causing it to move to the subsequent grain layers. Therefore, this treatment effect was accounted to the CTP quantification of each layer during a full treatment cycle starting when they enter the grain silo all the way to their exit. Mold reduction after treatment of each grain layer varied from 15 to 77%. No pattern was found between CTP increase and mold reduction increase.



**Figure 3** Ozone concentration at four different subsequent treated bottom grain layer in silo during semi-continuous counterflow ozonation treatment during several cycles of constant grain mass.

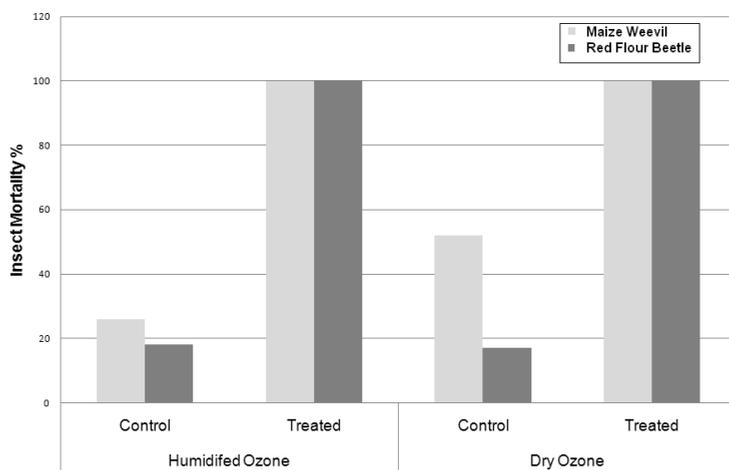
#### 3.2. Continuous ozonation flow treatment trial

During the treatment trial, the average residence time it took the grain sample to move from the bottom of the modified loading screw conveyor to its end (run) was 2 min for both applications of dry and humidified ozone. Based on theoretical calculations and ozone quantification in the distribution manifold, for each run of the grain samples through the system, the ozone concentration was 47,820 ppm.min. Therefore, the concentration-time product during each run was 95,640 ppm.min (47,820 ppm x 2 min).

Based on previous research work, the CTP for 100% insect mortality is 3,600 ppm.h that equals 216,000 ppm.min. At the continuous ozonation flow system 216,000 ppm.min is achieved at 4 min and 34 s between the second and third run. Therefore, it was determined that the 3 passes through the system was the minimum number of runs to try to achieve 100% insect mortality for both applications.

The average residence time for the 3-runs for both humidified and dry ozone showed a higher value of 6 min and  $31 \pm 0.10$  sec during dry ozone application compared to humidified ozone that had an average of 5 minutes and  $30 \pm 0.20$  sec. The control runs for humidified and dry ozone applications showed an average residence time of 5 min and  $21 \pm 0.22$  sec and 5 min and  $31 \pm 0.13$  sec, respectively.

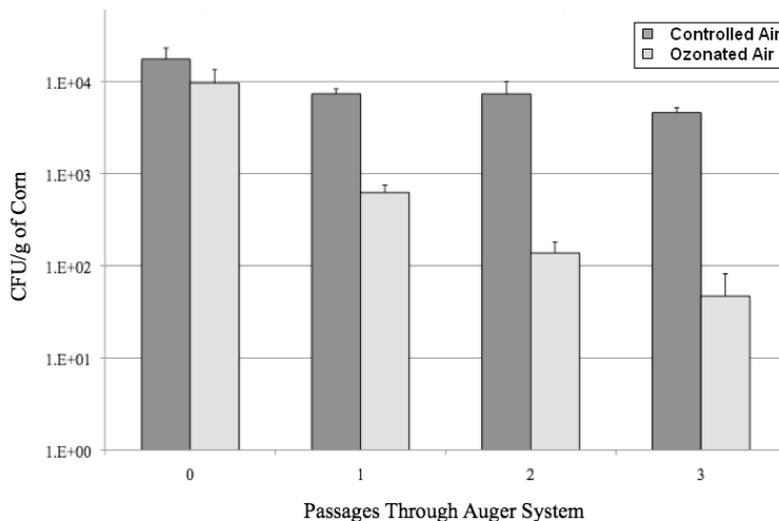
The insect mortality results for maize weevil and red flour beetle (Fig. 4) were 100% for the 3 runs tests using both humidified and dry ozone. The control 3-runs tests showed an average insect mortality of 18% for RFB and 30% for MW. The insect mortality in the control runs was caused by mechanical movement of the auger and the grain samples. The mold reduction results for the 3-runs treatments for both applications showed a considerable decreased from 11,646 to 6 CFU/g count for humidified ozone and 10,622 to 68 CFU/g count for dry ozone. Based on these results, it was shown that there is no difference between dry and humidified ozone application.



**Figure 4** Insect mortality (%) in bioassays of adult maize weevil and red flour beetle placed in grain samples for 3-runs treatments of continuous ozonation flow trial using humidified and dry ozone.

The insect mortality in the insect bioassays during 6, 4, and 2 min residence time for maize weevil resulted in 100, 82 and 64%, respectively. In the control runs for 6, 4, and 2 min residence time were 32, 24 and 7%, respectively. For red flour beetle, the insect mortality in the insect bioassays for 6, 4, and 2 min residence time resulted in 100, 57 and 79%, respectively. The insect mortality during the 4 min residence time treatment resulted in a lower value compared to the 2 min residence time treatment. No explanation can be found for this issue since in the 4 min residence time treatment the insect bioassays are exposed to almost double the ozone CTP than in the 2 min treatment. The control runs for 6, 4, and 2 min residence time, resulted in 18, 12, and 8%, respectively. Also, the insect mortality for 2 min residence time had a low value of 8%, therefore, it can be assumed that no other external factor caused the 1-run treatment to have a higher insect mortality than the 2-run treatment.

The mold reduction results (Fig. 5) showed a difference in CFU/g of maize mold count of almost 50% reduction between no treatment and ozone treatment. During treatment, the lowest mold count was obtained in the 3-runs treatment (6 min residence time) with a value lower than 5,000 CFU/g of maize due to the higher exposure time the grain was treated in the continuous ozonation flow system. Based on the standard deviation, there is no difference in mold count for the 1-run and 2-runs treatments or 2 and 4 min residence time. Therefore, it can be assumed that in order to have a more effective mold reduction, grain has to be exposed to at least a CTP of 300,000 ppm.min.



**Figure 5** Colony forming units per gram of maize (CFU/g) for mold count on grain for 3-runs, 2-runs and 1-run treatments and control run of continuous ozonation flow trial.

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